Response to Referee #2 (GMD-2023-22)

We Thank Reviewer for his/her constructive comments.

Responses to the comments:

Comments: The paper is quite ambitious, as it presents a novel approach (QDA - quantitative decoupling analysis) to quantify the effects of emissions, meteorology, chemical reactions and nonlinear interactions on $PM_{2.5}$ concentrations.

As the authors correctly state, in literature there are already existing approaches to perform this task, as IPR (integrated process rate), SAA (scenario analysis approach), FS (factor separation). On this last topic ... I have two main concerns on the current version of the paper

Reply: Thanks for this comment. I would reply in the following parts.

Comment 1: focus on pros and cons of the different approaches. Even if the authors provide some hints of the pros and cons of the approaches (section 2.1.4) still it is not clear to me why we need another approach (QDA) and why the existing ones are not sufficient. Please better explain this, and also provide a more schematic and syntetic view of pros and cons of the different approaches, i.e. also with a table or graphical view.

Reply: Thanks for this important comment. The differences between the QDA method and other mainstream methods in analyzing the effects of meteorology, chemistry and emission on the $PM_{2.5}$ are explained theoretically in Sect. 2.1.4. The gain of the QDA method compared to the IPR is straightforward that mainly exists in the resolving of the nonlinear effects among the different processes. As we illustrated in Sect. 3.3 and 3.4, we can resolve the changes of $PM_{2.5}$ into the pure effects of different processes (i.e. M, E, C) as well as their nonlinear interactions (i.e., EM, MC, CE and MCE) by using the QDA method, while the changes of $PM_{2.5}$ are only attributed to the effects of meteorology, chemistry and emission in the IPR method. The inclusion of the nonlinear effects in the QDA method, on the one hand, can overcome the problems of the no uniqueness of the results of IPR method which is dependent on the sequence of the different processes due to the nonlinearity of the atmospheric chemistry. On the other hand, it can provide us with more information on the state of atmosphere chemistry by analyzing the nonlinear effects. For example, the analysis of the EM results of the ammonium (Sect. 3.5.1) can help us identify whether there is a NH_3-rich or NH_3-poor condition.

The FS method is instructive to the development of the QDA method. However, in previous studies the FS method was mainly used in the analysis of the effects of model input parameters, such as the emission inventory and topography (Alpert et al., 1999; Tao et al., 2005), or the effects of specific physical variables, such as the synoptic-scale wind and the atmospheric moisture (Rotstein et al., 2021), which has not been used in the model processes. The QDA method takes the FS method as a reference and applies it to the analysis of the effects of meteorology, emission, chemistry as well as their interactions on the $PM_{2.5}$ concentrations. From this perspective, the QDA method is similar to the FS method but with different analysis objects and is applied within model steps.

The SAA has more than one choice of operation path, which leads to great uncertainty in the result. QDA method can overcome the problems of the SAA method in the dependence of the choices of fixed emission, which could yield consistent results of the effects of

meteorological and emission changes on the $PM_{2.5}$ variations.

Changes in the manuscript: lines 559-578.

Comment 2: also, I would like to see not only theoretically, but also in practice, what you gain using QDA instead of using other approaches. To do so, please apply, on the same data and episode, also other approaches (i.e. SAA, FS ...) to see if you really gain (and what you gain) on the results' quality and interpretation, using the QDA approach.

Reply: Thanks for this comment. Because FS may cause memory overflow or simulation error when it is used to calculate meteorological and chemical actions (turn off meteorology or chemistry for long-term simulation), only the differences between SAA and QDA are analyzed with a specific example in the revised manuscript.

To investigate the gains of the QDA method with regards to the SAA method, the SAA method was applied to the same cases in this study and compared with the QDA method. Commonly, the SAA method compares two cases that started at different times but lasted for the same duration. The concentration differences between these two cases can be divided into anthropogenic contribution and meteorological contribution. By keeping the emissions unchanged and changing the meteorological field in simulation, the contribution of meteorological changes to the $PM_{2.5}$ can be calculated, and the remaining change is the impact caused by the emissions. However, the results of this method are dependent on the choice of the emission. The different choices usually change the results of the SAA. In order to evaluate the proportion of anthropogenic and meteorological contributions in 19–27 February 2015 compared to the same period in 2014, the following scenarios are designed according to the SAA method (Table R1).

The changes in the concentrations of $PM_{2.5}$ between the two cases can be expressed as: $PM_{2.5\_2015base} - PM_{2.5\_2014base}$, and there are two paths to calculate the contribution of changes in emission and meteorological fields based on the SAA method:

Path 1: According to the Table R1, we can do it in these two steps:

$$2014base \rightarrow 2015\_emis2014 \rightarrow 2015base$$

The concentration changes for the two steps are:

$$\Delta MET1 = PM_{2.5\_2015\_emis2014} - PM_{2.5\_2014base}$$

$$\Delta ANT1 = PM_{2.5\_2015base} - PM_{2.5\_2015\_emis2014}$$

$\Delta MET1$ represents the amount of $PM_{2.5}$ concentration change due to meteorological changes, and $\Delta ANT1$ represents the anthropogenic contribution due to the emission changes.

Path 2: According to the Table R1, we can also do it in these two steps:

$$2014base \rightarrow 2014\_emis2015 \rightarrow 2015base$$

$$\Delta ANT2 = PM_{2.5\_2014\_emis2015} - PM_{2.5\_2014base}$$

$$\Delta MET2 = PM_{2.5\_2015base} - PM_{2.5\_2014\_emis2015}$$

In Table R3, the concentration differences can be decomposed into the contributions of anthropogenic (ΔANT), meteorological (ΔMET), and nonlinear effects ($\Delta COUP$), the sum of these three components represents the total change in $PM_{2.5}$ concentrations of 2015 compared to the same period of 2014. ΔMET is equal to the M mentioned above, ΔANT contains C, E and CE, and the rest of the nonlinear effects are classified as $\Delta COUP$. $\Delta COUP$ represents the nonlinear interaction between meteorological and anthropogenic effects and it can also be used as a range of uncertainty in pure meteorological and anthropogenic contributions. In

stage 2 and stage 3, reduction in $PM_{2.5}$ concentrations occurred in 2015 is more significant compared to 2014, and changes in meteorological conditions mainly contributed to it. Emissions in 2015 are lower than in 2014, which is the reason why ΔANT was negative in all stages, and the reduction in emissions will not only lead to the reduction in the direct contribution to $PM_{2.5}$, but also to the reduction in the precursors of chemical reactions, which will result in the less generation of secondary aerosol.

ΔMET2 and ΔANT2 represent the changes in $PM_{2.5}$ concentration due to meteorological changes and anthropogenic changes, respectively. And there is ΔMET1+ΔANT1 = ΔMET2+ΔANT2. It can be seen in the Table R2 that the results of anthropogenic and meteorological contributions of $PM_{2.5}$ obtained by different paths are not consistent, indicating that the results obtained by SAA have a large uncertainty. In stage 3, ΔMET1 valued -6.27 μg m^{-3} indicates that meteorological fields in 2015 were more unfavourable for $PM_{2.5}$ increases than in 2014, while ΔMET2 valued 0.3 μg m^{-3} reflects that the meteorological fields in 2015 were more favourable for $PM_{2.5}$ increases. The meteorological contributions of the two paths are even opposite. The anthropogenic contributions obtained by the two paths are also opposite in sign in stage 1 and 3, reflecting the uncertainties in the SAA method.

In Table R3, the concentration differences can be uniquely decomposed into the contributions of anthropogenic (ΔANT), meteorological (ΔMET), and nonlinear effects (ΔCOUP), the sum of these three components represents the total change in $PM_{2.5}$ concentrations of 2015 compared to the same period of 2014. ΔMET is equal to the M mentioned above, ΔANT contains C, E and CE, and the rest of the nonlinear effects are classified as ΔCOUP. ΔCOUP represents the nonlinear interaction between meteorological and anthropogenic effects and it can also be used as a range of uncertainty in pure meteorological and anthropogenic contributions. In stage 2 and stage 3, reduction in $PM_{2.5}$ concentrations occurred in 2015 is more significant compared to 2014, and changes in meteorological conditions mainly contributed to it. Emissions in 2015 are lower than in 2014, which is the reason why ΔANT was negative in all stages, and the reduction in emissions will not only lead to the reduction in primary $PM_{2.5}$, but also to the reduction in the precursors of chemical reactions, which will result in smaller formation of secondary aerosol.

In 2015 case, the difference between meteorological and anthropogenic contributions within the same stage is smaller than in 2014, so the variation in $PM_{2.5}$ is smaller in 2015. The meteorological fields can always provide clearance timely, so that the pollution produced by anthropogenic action cannot accumulate continuously, which is the main reason why there is no persistent heavy haze in 19–27 February 2015. Therefore, these results suggest that the QDA method can overcome the problems of the SAA method in the dependence of the choices of fixed emission, which could yield consistent results of the effects of meteorological and emission changes on the $PM_{2.5}$ variations.

Table R1. List of the scenario simulations

Scenario name	Time of meteorological field	Time of emission inventory

2014base	19–27 February 2014	19–27 February 2014
2015base	19–27 February 2015	19–27 February 2015
2014_emis2015	19–27 February 2014	19–27 February 2015
2015_emis2014	19–27 February 2015	19–27 February 2014

Table R2. Results of SAA for comparing vertical mean hourly concentration change of $PM_{2.5}$ in 19–27 February 2015 with the same period in 2014

	Path 1		Path 2	
Stage	ΔMET1 (μg m^{-3})	ΔANT1 (μg m^{-3})	ΔMET2 (μg m^{-3})	ΔANT2 (μg m^{-3})
Stage 1	142.95	-0.45	138.96	3.55
Stage 2	-204.00	0.31	-217.08	13.39
Stage 3	-6.27	0.63	0.30	-5.94
Stage 4	167.50	-0.68	174.02	-7.20

Table R3. Results of QDA for comparing vertical mean hourly concentration change of $PM_{2.5}$ in 19–27 February 2015 with the same period in 2014

Stage	ΔMET (μg m^{-3})	ΔANT (μg m^{-3})	ΔCOUP (μg m^{-3})
Stage 1	160.64	-7.25	-7.19
Stage 2	-178.10	-13.08	-13.42
Stage 3	58.89	-43.55	-20.63
Stage 4	166.17	-2.70	3.04

Notes: ΔMET represents changes in $PM_{2.5}$ concentrations due to changes in emission inventories, and also including changes in chemical reactions due to changes in emissions, ΔANT represents the change of $PM_{2.5}$ concentration caused by the change of meteorological fileds, ΔCOUP represents the change of $PM_{2.5}$ concentration caused by nonlinear effects.

Changes in the manuscript: lines 579-634.

Comment 3:

Minor comment

please move part of the Equations (section 2) in the supplementary material, so that the main concepts you propose remain in the main part of the manuscript, and the more technical part goes in the Annex.

Reply: Thanks for this comment. Part of equations in section 2 have been moved to the supplementary material in the revised manuscript as Eqs. (S1)–(S11).

Changes in the manuscript: lines 227-228, lines 294-295, lines 296-299.

References:

Alpert, P., Tsidulko, M., and Itzigsohn, D.: A shallow, short-lived meso-beta cyclone over the Gulf of Antalya, eastern Mediterranean, Tellus Ser. A-Dyn. Meteorol. Oceanol., 51, 249-262, 10.1034/j.1600-0870.1999.t01-2-00006.x, 1999.

Rotstein, M., Alpert, P., and Rostkier-Edelstein, D.: A Factor Separation Study of the Effect of Synoptic-Scale Wind, Atmospheric Moisture and of Their Synergy on the Diurnal Temperature Range During the Israeli Summer, J. Geophys. Res.-Atmos., 126, 18, 10.1029/2021jd034923, 2021.

Tao, Z. N., Larson, S. M., Williams, A., Caughey, M., and Wuebbles, D. J.: Area, mobile, and point source contributions to ground level ozone: a summer simulation across the continental USA, Atmos. Environ., 39, 1869-1877, 10.1016/j.atmosenv.2004.12.001, 2005.