

Dominant role of emission reduction in PM<sub>2.5</sub> air quality improvement in Beijing during 2013-2017: a model-based decomposition analysis by Jing Cheng et al.

### General Comments

The main purpose of this paper is the discussion of the reasons of recent rapid PM<sub>2.5</sub> decrease of Beijing, mainly by using the meteorological and emission sensitivities by chemical transport model. Based on several sensitivity simulations, this paper made a decomposition analysis framework to evaluate the impacts of local control policies, surrounding emission reductions and the meteorological changes on PM<sub>2.5</sub> abatement in Beijing during 2013-2017 and 2016-2017. This paper made the most of important sensitivity analysis and explains the relative contribution of meteorology (12%), local emission (65%) and regional emission (23%) for the reduction of PM<sub>2.5</sub> between 2017 and 2013. The results of detailed sensitivity analyses are useful for the understanding of PM<sub>2.5</sub> reduction and environmental policy. Their results are very much reasonable and important. However it is difficult to find scientific uniqueness of this paper. The current version of manuscript could be published as the Technical Note. It is necessary to add the more scientific discussion in order to be accepted as a research paper.

### **Response:**

**We thank the reviewer #1 for the constructive comments and address them as below.**

### Specific Comments

- 1) The paper mainly discussed the decreases of annual average PM<sub>2.5</sub> concentration. However as shown in Figure 4, the authors are specifying the decrease of chemical compositions (i.e., SO<sub>2</sub>, NO<sub>x</sub>), it is necessary to compare the aerosol chemical composition changes too. The under estimation of sulfate in winter are reported in many previous papers, the

comparison of sulfate between observation and model results are very much necessary.

**Response:** we add the validation of PM<sub>2.5</sub> compositions simulations in Sect.2.4.2 (the validation is conducted in terms of the absolute concentration values comparison and the temporal change comparison); the detailed comparison of the simulation and observations of PM<sub>2.5</sub> compositions are listed in SI, Table S4; the data source of observational PM<sub>2.5</sub> compositions is introduced in Sect.2.1 (SPARTAN observational PM<sub>2.5</sub> compositions data was the most complete public data we could found, however with certain temporal discontinuities. This may not be able to support the trend analysis, but can used for period validation. Besides this observational data set, we also collected some trends observational analysis in reported research (Shao et al., 2018) to verify the temporal changes of simulated PM<sub>2.5</sub> compositions).

As Table S4 showed, the simulation of sulfate basically agrees with the observation, even with slight overestimation in winter. On the one hand, a missing heterogeneous chemistry mechanism is revised in our model based on our previous work (Zheng et al., 2015), which can improve the sulfate simulation in pollution events. On the other hand, along with the effective SO<sub>2</sub> emission control measures in Beijing in recent years, the SO<sub>4</sub><sup>2-</sup> was basically no longer the key contributor leading to heavy pollution, while the nitrate-driven haze pollution has become more dominate, especially in the summertime (Li et al., 2018). Additionally, the estimation of SO<sub>2</sub> emission might be overestimated, for the obvious overestimation of SO<sub>2</sub> simulation (SI, Table S3).

We also add an analysis about the aerosol chemical composition changes in Sect.3.4.1 as follows, and the variation trends of simulated PM<sub>2.5</sub> compositions can be found in SI, Figure S6.

**Added/rewritten part in Sect.2.1:** To evaluate the accuracy of PM<sub>2.5</sub> components simulation, we collected the PM<sub>2.5</sub> components data from the Surface PARTiculate mAtter Network (SPARTAN, [www.spartan-network.org](http://www.spartan-network.org)). SPARTAN is a global, long-term project that observed and analyzed the particulate matter mass, water-soluble ions, black carbon and

metals since 2013 (Snider et al., 2015). Major SPARTN measurements include the Air Photon 3-wavelength integrating nephelometer and the Air Photon SS4i automated air sampler. SPARTAN monitors are located in nearly 20 highly populated regions in the world, such as Beijing, Hanoi, Singapore, Dhaka, Pretoria, Toronto, Bondville. Detailed measurements and sites information can be found in Snider's research (Snider et al., 2015; Snider et al., 2016). In this research, we used the reconstructed PM<sub>2.5</sub> speciation data in Beijing Site (located in the Department of Earth System Science, Tsinghua University) (<https://www.spartan-network.org/beijing-china>) to validate the model simulation results of PM<sub>2.5</sub> compositions, and to verify the variation of PM<sub>2.5</sub> compositions in Beijing during 2013-2017. Additionally, we collected the observational PM<sub>2.5</sub> compositions from the reported research (Shao et al., 2018), to verify the simulated variations of PM<sub>2.5</sub> compositions in CMAQ model. In Shao's research, they observed and analyzed the multi-size PM samples at an urban site on Beijing Normal University, and compared the chemical compositions changes from January 2013 to 2016-2017 winter in Beijing.

**Added/rewritten part in Sect.2.4.2:** Besides PM<sub>2.5</sub> concentrations, we also evaluated the simulated PM<sub>2.5</sub> compositions. We compared the PM<sub>2.5</sub> compositions of the observations from Beijing Site, SPARTAN and the simulations from the same grid, which can be found in SI (Table S4). Generally speaking, secondary inorganic aerosol (SIA) was overestimated in most periods (0.1%-56.6%), especially for the nitrate (NO<sub>3</sub><sup>-</sup>) simulation, which overestimated 6.56%-89.9%. Organic matter (OM) was under estimated in most periods, with the underestimation range of (-3.0%) - (-41.8%). This might be caused by the missing mechanism and insufficient simulation of secondary organic aerosols (SOA) formulations in CMAQ model. The NMB of simulated and observed black carbon (BC) varied from (-52.9%) – (38.2%). Similar evaluation results can be found in Weagle's research, that he compared the GEOS-Chem simulation with SPARTAN observation data, with an average 45.7% overestimation of SIA, -58.9% underestimation of BC, and -19.2% underestimation of OM. However, the variation trends of simulated PM<sub>2.5</sub> compositions were basically consistent with the SPARTAN data, both have the remarkable decrement in OM and increment in NO<sub>3</sub><sup>-</sup>. In Shao's observational results, that SO<sub>4</sub><sup>2-</sup> proportion in 2016-2017 winter in Beijing reduced by 11% compared with January,2013; while NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> proportions increased by 77.9% and 47.3% (Shao et al., 2018). And in our research, the relative change ratio of SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> proportions in Beijing from January, 2013 to 2016-2017 winter were -29.1%, 89.2% and 11.7% respectively. In general. The simulated compositions basically captured the

variations in observation results, which can support the reasonable analyses of the chemical composition changes.

**Added/rewritten part in Sect.3.4.1:** Although there was a steady decline in  $PM_{2.5}$  concentrations of Beijing during 2013-2017, the trends of  $PM_{2.5}$  compositions varied differently. The simulation results of base cases (which adopted the real meteorology and emissions of each year) showed that the sulfate ( $SO_4^{2-}$ ) and organic matter (OM) were dominate species for the decline in  $PM_{2.5}$  concentrations during 2013-2017, with the decrement of  $7.5 \mu g m^{-3}$  (56.6%) and  $9.6 \mu g m^{-3}$  (40.5%) respectively. The contribution of  $SO_4^{2-}$  to the total  $PM_{2.5}$  also decreased obviously, from 15.3% in 2013 to 10.7% in 2017; and OM proportion decreased from 27.5% in 2013 to 26.5% in 2017. The rapid decrement of  $SO_4^{2-}$  was consistent with the remarkable  $SO_2$  emission reductions in Beijing during 2013-2017. Along with the effective  $SO_2$  emission control measures, the  $SO_4^{2-}$  was basically no longer the key contributor leading to heavy pollution in Beijing while the nitrate-driven haze pollution has become more dominate in Beijing in recent years, especially in the summertime (Li et al., 2018). The decrement of OM was mainly caused by the prominent emission reductions of primary organic carbon (mainly come from the residential burning and other coal combustion sources). VOCs emission reductions also contributed to the OM decreasing, however, due to the missing mechanism and insufficient simulation of secondary organic aerosols (SOA) formulations in CMAQ model, the contributions of VOCs emission control might be underestimated. In contrast, nitrate ( $NO_3^-$ ) increased in 2014-2016, and kept basically the same concentration level in 2017 ( $10.4 \mu g m^{-3}$ ) as 2013 ( $10.9 \mu g m^{-3}$ ). However, the contribution of  $NO_3^-$  to the total  $PM_{2.5}$  increased a lot, from 12.7% in 2013 to 19.4% in 2017. The specific concentration and proportion trends of  $PM_{2.5}$  concentrations can be found in SI, Table S6.

- 2) Another important point is the OC (we found that model result are usually under estimated), so the detailed examinations of model reproductively of OC are necessary in order to discuss the emission change in VOC.

**Response: please refer to the response for Specific Comments (1).**

- 3) The authors show the very good agreement of PM<sub>2.5</sub> reduction between observation and model results. It is usually difficult to have such good agreement. It is necessary to discuss the detailed reasons why model results are so good agreement from the view point of emission inventory, WRF model, model horizontal resolution, and CMAQ itself performance.

**Response:** In our simulation, a better emission inventory of Beijing (described in Sect.2.2), with more accurate e spatial distribution and emission source allocation, was adopted in our simulation, which can largely improve the air quality modelling, especially when modelling with finer resolutions (Zheng et al., 2017). Meanwhile, analysis nudging, observational nudging and soil nudging were adopted in WRF simulation, which improved the meteorology modelling (SI, Table S2 (a)-(f)). The statistic of WRF simulation results showed a better performance. Additionally, three nested domains were designed and the horizontal resolution of the third domain was 4km × 4km. The higher resolution enhanced the advantage and accuracy of BJ-EI; meanwhile the simulation of finer grids would create higher spatial accuracy and better agree with the observational data. However, the model was not entirely perfect. On the one hand, certain biases existed in the simulated and observational concentrations (such as the underestimation of PM<sub>2.5</sub>, O<sub>3</sub> and the overestimation of SO<sub>2</sub> (SI, Table S3(b)), especially in some heavy pollution episodes. And the biases became even larger in the PM<sub>2.5</sub> compositions comparisons (SI, Table S4). On the other hand, the presented validation analysis was the average results from 12 observation stations in Beijing. The simulation (both the reductions and absolute concentrations) of different grids (where different observation stations located) also presented different performance, which can be seen from the certain ME and NME statistics (SI, Table S3, Table S4). Generally speaking, the better emission inventory and higher resolutions in this study led to a better model performance, however certain biases and uncertainties existed due to the missing mechanism in the model itself and some other limitations.

- 4) Zero-out emission sensitivity is used in this study by assuming the linearity. PM<sub>2.5</sub> formation is usually nonlinear, so it is necessary why the authors are using zero-out method.

**Response: we add a discussion part (Sect.3.5.1) to quantify the extra non-linearity effects of the zero-out approach in our study; meanwhile, we also explained the reason why we used zero-out approach in Sect.3.5.1 as follows.**

**Added/rewritten part in Sect.3.5.1:** Although various methods have been developed to quantify the source of PM<sub>2.5</sub> and evaluate their contributions, such as receptor-based methods (like CMB and PMF), trajectory-based methods (like PSCF and EEI), source-oriented methods (like CAMx-PSAT and CMAQ-ISAM)) (Li et al, 2015), they can hardly consider the meteorology and emission changes simultaneously. Therefore, the zero-out approach might be a better choice to attribute the contribution of local and regional emission control as well as meteorology changes under one complete decomposition framework. The zero-out method is also widely used in estimating the contribution of air pollution sources (Lelieveld et al., 2015; Han et al., 2016; Baker et al., 2016; Zhang et al., 2017; Zhang et al., 2017; Ni et al., 2018).

However, the response of PM<sub>2.5</sub> formulation is not linear to the meteorology and emission changes; thus, the zero-out approach would introduce extra bias in research. The non-linear effects of the analyse period of 2013-2017 could be evaluated by the following equation (Zhang et al., 2017).

$$\text{Bias} = (\text{SCon}(M) + \text{SCon}(S) + \sum_{i=1}^7 \text{SCon}(pi)) - (\text{SPM}_{2.5}(E_{L13S13}M_{13}) - \text{SPM}_{2.5}(E_{L17S17}M_{17})) \quad (7)$$

Where  $\text{SPM}_{2.5}(E_{L13S13}M_{13})$  and  $\text{SPM}_{2.5}(E_{L17S17}M_{17})$  represent the direct simulated PM<sub>2.5</sub> concentration of base case in 2013 and 2017. The balance of their values is the actual PM<sub>2.5</sub> decrement during 2013-2017 under the mixed impacts of meteorology change, regional and local emission reductions. The sum of  $\text{SCon}(M)$ ,  $\text{SCon}(S)$  and  $\sum_{i=1}^7 \text{SCon}(pi)$  represents a linear result of all contributors during this period. The extra bias can be estimated as the difference between the linear addition and the actual decrement. According to equation (7), we estimated biases in the analyse of 2013-2017 were 1.4  $\mu\text{g m}^{-3}$ , accounting for 4.3%. Similarly, the absolute and relative biases in the analysis of 2016-2017 were estimated as -0.6  $\mu\text{g m}^{-3}$  and -3.6%. Both indicated the non-linear effects are relatively small and acceptable.

- 5) Section 2.3: Although we can follow by Zheng et al. (2018) to understand the emission inventory, it should be noted what is the major “updated”. Especially, the inventory is named as “MEIC”; however, we noticed the emission amounts described in Zheng et al. (2018) and Li, M. (2017) are different. Does this mean “updated”? Taking into account the importance of emission inventory, more careful explanations and descriptions are needed here for the traceability of this kind of study.

**Response: The MEIC model is developed by Tsinghua University, and keeps continuous updating. Li, M. (2017) focused on the year of 2008, 2010 and 2012; while Zheng et al. (2018) focused on the periods of 2010-2017, discussing the temporal emission changes in China from 2010-2017. We add some detailed information about Zheng’s work to explain the major updated specifics in MEIC model in Sect.2.3 as follows; and a brief introduction of MEIC is also added.**

**Added/rewritten part in Sect.2.3:** The air pollutant emission inventory of Beijing’s surrounding regions (including Tianjin, Hebei, Henan, Shandong, Shanxi, Inner Mongolia) for the period of 2013-2017 was obtained from the MEIC model. MEIC is a bottom-up emission inventory model developed for China by Tsinghua University, covering 31 provinces in China and the year range of 1990 up to now. More than 700 emission sources were developed in MEIC model. The methodologies and data on which the MEIC is based, as well as the continues updating process of pollutants emission factors, have been introduced in our previous studies (Zhang et al 2007; Liu et al., 2010; Lei et al., 2011; Li et al., 2014; Shen et al., 2014; Hong et al., 2017; Qi et al., 2017; Zheng et al., 2017; Li et al., 2017a; Li et al., 2017b). For the updated emission inventory and emission reductions of the surrounding regions, we referee to our previous work (Zheng et al., 2018). In their work, Zheng et al collected the latest Chinese Energy Statistics data from National Bureau of Statistics, the industrial production and technology penetration data, and the unpublished data from the Ministry of Ecology and Environment. Then they estimated and updated China’s anthropogenic emissions from 2010 to 2017 under the framework of MEIC model. Particularly, they calibrated the emission calculation parameters (such as technology penetrations and removal efficiencies) in major sectors, such as power, cement, steel, iron, of each province based on emission control policies. Based on their updated China

anthropogenic emission inventory, we collected and analyzed the detailed sectoral emission reductions and trends of Beijing's surrounding regions during 2013-2017.

- 6) Section 2.4.1: Model descriptions are insufficient. Model calculation was conducted after only 10 days spin-up. How does the 3 years WRF simulation perform? i.e., with or without FDDA? Need more detailed descriptions.

**Response: we rewrite and add some detailed descriptions about the WRF-CMAQ model configurations in Sect.2.4.1 (as follows). In our study, we made a complete base simulation of 2013-2017 and obtained a consecutive five-year WRF and CMAQ simulation results. The 10 days spin-up was adopted to 18 sensitivity experiments. Analysis nudging, observational nudging and soil nudging were adopted, and FDDA data was from the U.S. National Centers for Environmental Prediction (NCEP, <http://rda.ucar.edu/datasets/>), Automated Data Processing surface (ds461.0) and upper (ds351.0) air data. The nudging-related configurations in WRF model were referred to our previous work (Zheng et al., 2015) and presented a good performance in meteorology modelling of North China.**

**Added/rewritten part in Sect.2.4.1:** For the WRF model configuration, we chose the New Goddard scheme (Chou et al., 1998) and RRTM (Mlawer et al., 1997) for shortwave and longwave radiation options, the Kain-Fritsch cloud parameterization (version 2, Kain, 2004), the ACM2 PBL scheme (Pleim, 2007), the Pleim-Xiu land-surface scheme (Xiu and Pleim, 2000) and WSM6 cloud microphysics (Hong and Lim, 2006). Analysis nudging, observational nudging and soil nudging were adopted, and FDDA data was from the U.S. National Centers for Environmental Prediction (NCEP, <http://rda.ucar.edu/datasets/>), Automated Data Processing surface (ds461.0) and upper (ds351.0) air data. The meteorological initial and boundary conditions were derived from the final analysis data (FNL). We made a continuous meteorology simulation during 2013-2017, with a ten-days spin-up before this period.



- 7) Does CMAQ study include the Asian dust? If yes, need discussion of model accuracy and problems.

**Response: Asian dust was included in this study. We add the dust emission descriptions in Sect.2.4.1 as follows; and we also add a discussion about the relevant simulation results in Sect.2.4.2.**

**Added/rewritten part in Sect.2.4.1:** For the dust emission, bare lands dust was calculated by the in-line windblown dust in CMAQ model. As 2.2 section described, other dust source, such as road dust and construction dust were added in BJ-EI, while missed in MEIC model. It brought uncertainty of air quality simulation, especially for the PM<sub>10</sub> simulation in other regions.

**Added/rewritten part in Sect.2.4.2:** The added fugitive dust emission (road and construction dust) in BJ-EI has improved the PM<sub>10</sub> simulation of Beijing, with an overestimation range of 4% - 12% (Table S3 (b)). However, the PM<sub>10</sub> simulation of other cities in the third domain were underestimated (-8%) - (34%), especially in some heavy industry cities as Tangshan (-34%), Baoding (-25%), and Handan (-29%). This might be attributed to lack of construction dust emissions in these regions, as well as the uncertainty of the dust model (Todd et al., 2008; Foroutan et al., 2018). Similar methods and phenomenon were also applied and reported in previous researches (Wang et al., 2010). Although it might introduce the uncertainty of simulation; given that our research was focused on the attribution analyses of anthropogenic emission changes in Beijing, this uncertainty was relatively small.

- 8) Emission sensitivity study was conducted by each emission sector base. It is necessary to include the discussion of the accuracy (or error bars) of emission estimate for each sector base.

**Response: we add a discussion part to describe the uncertainty of emission estimations in Sect.3.5.2 (as follows). We first represent the quantified error bars of MEIC model (Zhang et al., 2009) and the uncertainty of regional emission estimations (Zheng et al., 2018) referred to our previous work. Then based on**

**Zheng's method, we discussed the uncertainty of BJ-EI (the local emission inventory of Beijing) (Sect.3.5.2; SI, Figure S5). Finally, we discussed the uncertainty in the estimation of measure-based emission reductions.**

**Added/rewritten part in Sect.3.5.2:** The incomplete research and investigation of activity rates, emission factors, and removal efficiencies would introduce uncertainties in estimating the emission trends and magnitudes, as well as the measure-based emission reductions.

Based on our previous work, the uncertainty of MEIC was estimated to be  $\pm 12\%$  for SO<sub>2</sub>,  $\pm 31\%$  for NO<sub>x</sub>,  $\pm 70\%$  for CO,  $\pm 68\%$  for VOCs,  $\pm 130\%$  for PM<sub>2.5</sub>,  $\pm 208\%$  for BC and  $\pm 258\%$  for OC (Zhang et al., 2009). The larger uncertainty of BC and OC are mostly because their major emitters are much more scattered and harder to investigate or quantify, such as heavy diesel vehicle and residential burning. The uncertainty of updated emission inventory of China during 2010-2017 was evaluated in Zheng's work by comparing the emissions with observations (Zheng et al., 2018), and reported a good agreement.

Similarly, we discussed the uncertainty of emission trends and relative change ratios in BJ-EI. Ground-based, satellite-based observational data and the estimated emissions from BJ-EI were all normalized to the base year, 2013. We compared the observational concentration trends of major pollutants with their precursor emissions (SI, Figure S5). A good agreement was found in the trends of SO<sub>2</sub> emission, OMI SO<sub>2</sub> column and surface observational SO<sub>2</sub> concentration, with a decreasing ratio of 83.6%, 77.6%, 69.8% respectively (SI, Figure S5(a)), indicating a relatively small uncertainty in SO<sub>2</sub> emission estimation. Surface concentration trend during 2016-2017 became flatter than OMI SO<sub>2</sub> columns trend, which might be partly owing to the relatively less surface SO<sub>2</sub> emission reductions while a more significant high-stack emission reductions in local and surrounding areas. The NO<sub>x</sub> emission trend was basically consistent with the variation in NO<sub>2</sub> tropospheric vertical column, decreasing 43.2% and 40.3% respectively, but both lower than the surface NO<sub>2</sub> concentration trend (SI, Figure S5(b)). This phenomenon might be caused by the meteorology impacts (Uno et al., 2007), chemical reactions of nitrous oxides (Valin et al., 2011), and the overestimation of surface NO<sub>2</sub> observations (Lamsal et al., 2010). Satellite-derived and surface observational PM<sub>2.5</sub> concentrations decreased 36.0% and 35.2% respectively during 2013-2017 in Beijing, both agreed with the trends of primary PM<sub>2.5</sub> and precursor emissions (SI, Figure S5(c)). Among all precursors, the decreasing rate of SO<sub>2</sub> was more significant and rapid than that of NO<sub>x</sub> and HN<sub>3</sub>, which was consistent with the simulation results that the proportion of sulfate in PM<sub>2.5</sub> decreased obviously while the contributions of nitrate and ammonium increased (SI, Figure

S4). The decrement of VOCs emissions contributed to PM<sub>2.5</sub> abatements by decreasing OM. Similar phenomenon was also reported in previous researches (Shao et al., 2018). In general, the relatively good coherence of emission and observation variations indicated that the BJ-EI basically well quantified the actual emission trends and variations in Beijing during 2013-2017.

Estimation of measure-based emission reductions is another major aspect of introducing uncertainty. However, the uncertainty of this aspect is hard to quantify. Emission control measures can have independent or integrated impacts on activity rate, emission factor, technology evolution, and end-of-pipe removal efficiency, which are all sensitive to emission calculation and remaining large uncertainty. *Coal-fired boilers control* was the most explicit control policy to quantify, for the detailed and accurate information of unit-based power plants and facility-based boilers. The amount of eliminated coal, newly increased other clean energy, the evolutions of emission factor and removal efficiency of each boiler were collected sufficiently, which can largely lower the uncertainty of the reduction estimation. Similar to *Coal-fired boilers control*, *Improve end-of-pipe control* mainly focused on the heavy-polluted manufacturing factories and gas/oil-fired boilers. The facility-based information and the accurate eliminated capacities make the uncertainty relatively small. As for the policy of *Clean fuels in residential sector*, the reduced residential coal use, the eliminated traditional biofuels were collected at a county-level, thus the uncertainty of activity rate estimation was relatively small. However, along with the promotion of coal quality (such as the lower sulfur and ash content) and the evolution of domestic burning equipment, the improvement of emission factors was hard to estimate, especially in the rural areas, which would introduce large uncertainty. Major limitations of estimating *Optimize industrial structure* reductions came from the elimination of small, clustered and polluting factories. Different from high-polluted enterprises, the specific information of these scattered factories was hard to investigate. Although we knew a total of 11,000 such factories were phased out during 2013-2017, the activity rate, emission factor or end-of-pipe control information were ambiguous, which would easily lead to the underestimation or overestimation of reductions from this sub-measure. *Integrated treatment of VOCs* included the VOCs control of chemical industry and solvent use sectors. Yan Shan company is the only chemical factory in Beijing, and the specific information of this factory made the estimation more reasonable. However, investigations for the amounts of solvent used were limited; meanwhile, the lack of emission factor measurements for various solvent-use-related sources also introduced large uncertainty (Li et al., 2017a). Reductions from *fugitive dust control* might be the most difficult one to

estimate, with the larger uncertainty. This policy type contained the control of road dust and construction dust. Due to the lack of measurements for the real-time traffic flow, threshold friction velocities, the efficiency of road cleaning process and other key parameters for emission calculation, we estimated the emission reductions from this sub-measure by the improvements of cleaning process adopted ratio and the various road areas, which might not totally reflect the actual emission change of road dust. For the construction dust estimation, although we collected the information of each construction site and stock yard in Beijing, the indefinite emission process and factors would also create uncertainties for estimation. In general, emission reductions from the policies which focused on non-point and scattered emission sources, such as road dust, small and clustered factories, various solvent use sources, are more difficult to quantify, and would cause larger uncertainties. Additional detailed information and real-world measurements might help to lower these uncertainties.

- 9) Section 2.5 is unclear. The description of model sensitivity has to be rewrite. Equations of (1) – (3) are unclear. I think Equations of (2) and (3) are not NORMALIZED RESULTS.

**Response: we rewrite the Sect.2.5 (Scenario design and decomposition analysis) as follows:**

**Added/rewritten part in Sect.2.5:** All scenario cases were labelled as  $E_{Lisj}M_k$ .  $M_k(k)$  represents the metalogical period the case adopted and  $E_{Lisj}(i, j)$  represents the emission period. Total emission inventories of China consisted of two parts, that the BJ-EI from BMEMC and the regional (all parts of China except for Beijing) emission inventories from MEIC model. The adopted emission period of these two parts were labelled as  $Li(i)$  and  $Sj(j)$  respectively.

$E_{L13S13}M_{13}$ ,  $E_{L16S16}M_{16}$ , and  $E_{L17S17}M_{17}$  were three base cases and driven by the actual emission inventories and meteorology of 2013, 2016 and 2017, respectively, to reproduce the air quality of the corresponding year.  $E_{L17S17}M_{13}$  and  $E_{L17S17}M_{16}$  were designed to investigate the impact of meteorology. These two cases were driven by varying meteorological conditions (meteorology of 2013 and 2016, respectively) and the same emission inventory (for the year 2017).  $E_{L17S13}M_{17}$  and  $E_{L17S16}M_{17}$  were designed to quantify the impact of surrounding emission reduction during 2013-2017 and 2016-2017. In these two cases, the emission inventory of Beijing was set to the 2017 level, while the regional emission inventory was set to the 2013 and 2016 levels, respectively.

Another fourteen simulations were designed to quantify the air quality improvements contributed by seven types of local control policies during two periods. Cases for 2013-2017 and 2016-2017 were labelled as  $E_{L_{pi}S17}M_{17}$  and  $E_{L_{qi}S17}M_{17}$  respectively, where  $i$  represents the number of each policy (described and listed in Table 1). The meteorological conditions and regional emission inventories of these fourteen cases were set to 2017. For each simulation, emission reduction introduced by the corresponding policy type and adopting period was added to the 2017 baseline, equivalent of “turning off” this type of policy during this period. And then the derived emission inventory was applied to drive the corresponding air quality modelling. A linear additive relationship was assumed among all contributors to perform a decomposition analysis, and the simulated contributions of all sensitivity cases were then normalized by the difference in observed  $PM_{2.5}$  concentrations from 2013-2017 and 2016-2017. The normalization process of 2013-2017 period were calculated by the following equations, while the simulated results for period of 2016-2017 can be normalized with the similar process.

$$SCon(M) = SPM_{2.5}(E_{L17S17}M_{13}) - SPM_{2.5}(E_{L17S17}M_{17}) \quad (1)$$

$$SCon(S) = SPM_{2.5}(E_{L17S13}M_{17}) - SPM_{2.5}(E_{L17S17}M_{17}) \quad (2)$$

$$SCon(pi) = SPM_{2.5}(E_{L_{pi}S17}M_{17}) - SPM_{2.5}(E_{L17S17}M_{17}) \quad (3)$$

$$NCon(M) = (PM_{2.5OBS2013} - PM_{2.5OBS2017}) \times \frac{SCon(M)}{SCon(M) + SCon(S) + \sum_{i=1}^7 SCon(pi)} \quad (4)$$

$$NCon(S) = (PM_{2.5OBS2013} - PM_{2.5OBS2017}) \times \frac{SCon(S)}{SCon(M) + SCon(S) + \sum_{i=1}^7 SCon(pi)} \quad (5)$$

$$NCon(pi) = (PM_{2.5OBS2013} - PM_{2.5OBS2017}) \times \frac{SCon(pi)}{SCon(M) + SCon(S) + \sum_{i=1}^7 SCon(pi)} \quad (6)$$

where  $SCon(M)$  represents the simulated contribution of meteorology change during 2013-2017, which equals the balance of simulated  $PM_{2.5}$  ( $\mu g m^{-3}$ ) from case  $E_{L17S17}M_{13}$  and case  $E_{L17S17}M_{17}$ . Similarly,  $SCon(M)$  and  $SCon(pi)$  represent the simulated contribution of regional emission reductions and each local control policy type.  $NCon(M)$  represents the normalized contribution of meteorology change during 2013-2017, which equals the product of the observational  $PM_{2.5}$  balance (from 2013-2017) and the proportion of simulated meteorology contribution (in the simulated contributions of all factors). Similarly,  $NCon(M)$  and  $NCon(pi)$  represent the normalized contribution of regional emission reductions and each local control policy type.

10) Section 3.2 Without the model evaluation from 2013 to 2017, it is hard to discuss the source attribution results by model. From the current

manuscript, we can only find time-series on 2013, 2016, and 2017 for PM<sub>2.5</sub> and statistic evaluation only on 2017. The model evaluation is inadequate at the current manuscript. For example, Figure 3 and Figure S3 can be presented in the same form for model. See also my minor comments 4) for O<sub>3</sub> performance.

**Response: we add the consecutive evaluation (2013-2017) of WRF and CMAQ simulation results in SI (Table S2 (a)-(f) for WRF validation; Table S3 (a) for monthly descriptive statistic of PM<sub>2.5</sub> simulation; Table S4 for validation of PM<sub>2.5</sub> compositions; and Table S3 (b) for annually statistic of six major pollutants). Relative analysis is also added in Sect.2.4.2 (as follows).**

**Added/rewritten part in Sect.2.4.2:** The time series and evaluation results indicated that the CMAQ model and simulation results in this work can relatively well reproduce the temporal and spatial distribution of air pollutants in Beijing and its surroundings. As for the simulated PM<sub>2.5</sub> of 2017, the monthly Corr of PM<sub>2.5</sub> concentrations varied from 0.53 (in May) to 0.89 (in October), and the annual Corr of PM<sub>2.5</sub> concentrations varied from 0.65 (in 2016) to 0.81 (in 2014). The NMB and NME of monthly PM<sub>2.5</sub> simulations were within  $\pm 45\%$  and  $\pm 55\%$ , respectively. According to the observation data, the annual average PM<sub>2.5</sub> concentrations in Beijing decreased by  $31.5 \mu\text{g m}^{-3}$  from 2013 to 2017, while the simulated PM<sub>2.5</sub> decreased by  $32.8 \mu\text{g m}^{-3}$  (Table 2). Compared with 2016, the observed and simulated PM<sub>2.5</sub> decreased by 14.9 and  $16.6 \mu\text{g m}^{-3}$ , respectively (Table 2). The evaluation results suggested that the modelling system in this work can be used to quantify and analyses the attribution of PM<sub>2.5</sub> mitigations in Beijing. As for the simulation results of other pollutants in Beijing, the Corr varied from 0.61-0.74 for SO<sub>2</sub>, 0.59-0.68 for NO<sub>2</sub>, 0.62-0.78 for CO, 0.64-0.74 for O<sub>3</sub>, 0.62-0.74 for PM<sub>10</sub> (Table S3 (b)), which was acceptable for the research. The SO<sub>2</sub> simulation was overestimated in the five years, especially during 2013-2015, which indicated the SO<sub>2</sub> emission in BJ-EI might be higher than the reality. The added fugitive dust emission (road and construction dust) in BJ-EI has improved the PM<sub>10</sub> simulation of Beijing obviously, with an overestimation range of 4% - 12% (Table S3 (b)). However, the PM<sub>10</sub> simulation of other cities in the third domain were underestimated (-8%) - (34%), especially in some heavy industry cities as Tangshan (-34%), Baoding (-25%), and Handan (-29%). This might be attributed to lack of construction and road dust emissions in these regions, as well as the uncertainty of the dust model (Todd et al., 2008; Foroutan et al., 2018). It might introduce

the uncertainty of simulation, but given that our research was focused on the attribution analyses of anthropogenic emission changes in Beijing, this uncertainty was relatively small. The O<sub>3</sub> was underestimated in this WRF-CMAQ model system, with the range of (-8.3%) - (-22.6%). The rough vertical layers, the underestimation of nature source emissions, the defect of upper boundary simulation in regional model and the uncertainties of VOCs emission inventories might all lead this underestimation.

- 11) The results shown here should be interpreted in depth. On 2013, especially the peak on January, model sometimes overestimated observed PM<sub>2.5</sub>. However, model simulated same level or sometimes underestimated high concentrations during winter on 2016 and 2017. Actually, the model negative-bias is larger in 2017 compared to 2013 (Table 2). Therefore, the source attribution results based on scenario analysis adopted in this study can be strongly reflected by emission variation rather than the observed facts.

**Response: more detailed validation analysis is added in Sect.2.4.2, SI, Table S2-Table S4 (please refer to the response for Specific Comments (1), (3), (10), and Minor Comments (4)). The instability of high concentrations simulation might largely attribute to the variation of emission during 2013-2017. In 2013, all pollutants basically emitted at a high level, while decreased remarkably in 2017. The emission change and the relevant response in CMAQ model, as well as the impacts of meteorology, might lead to this instability.**

**As this comment pointed out, the source attribution results based on scenario analysis adopted in this study can be strongly reflected by emission variation. Therefore, we add a discussion part in Sect.3.5.3 to analysis the uncertainty of emission estimation.**

- 12) Figures 5 and 9 are unfriendly. It need more detailed explanations or improve the presentation of figures. The basic information is same as

Figure 10, so it might be better to modify Figures 5 and 9 into Figure 10 format.

**Response: we updated the Figure 5 and 9 based on Figure 10 format, and add some extra explanation.**

13) Section 3.3: The discussion in this section needs relevant references.

**Response: We add some relevant references in this part. Additionally, the discussion of this section is developed based on Zheng's work (Zheng et al., 2018). In their work, Zheng et al updated China's emission during 2010-2017 based on latest national statistics and relevant control policies. The mainly updates are briefly introduced in Sect. 2.3, and the relevant uncertainty is added in Sect.3.5.2. Based on this China emission inventory, we extract the emissions of Beijing's regional areas, including Tianjin, Hebei, Henan, Shandong, Shanxi and Inner Mongolia, and then analyze the emission variation of these areas during 2013-2017. The emission results are consistent with Zheng's work, while the relevant policy and attribution analysis is based on national clean air action plan.**

### Minor Comments

1) It have been reported that WRF should be updated version 3.9.0.1 or later for the upgraded NCEP dataset after 12 UTC, 19 July 2017.

<http://www2.mmm.ucar.edu/wrf/users/wpsv3.9/known-prob-3.9.html> The used version is 3.8, but how did the authors solve this problem?

**Response: we used the WRF version of 3.8 and the WPS version of 3.9.1. Given that the forward compatibility, the results of WPS 3.9.1 can be used to drive WRF3.8 simulations.**

2) What is the horizontal grid resolution in second domain?



**Response: the horizontal grid resolution in second domain is 12km × 12km, we also add this description in Sect.2.4.1.**

- 3) What is the lateral boundary condition for first domain? It will be taken from global chemical transport model, but did the global model consider year-to-year emission variation? If not, how can we conclude the importance of global-scale impacts on the air quality in China?

**Response: the chemical initial and boundary conditions (ICBCs) of the first domain were interpolated from the output of GEOS-Chem model (Bay et al., 2001; Geng et al., 2015). The year-to-year global emission variation was provided by the emission inventories and scale factors from GEOS-Chem Shared Data Directories (<http://acmg.seas.harvard.edu/geos/doc/man/>). However, given that our research is focused more on Beijing, the nation-scale impacts, especially the impacts of emission reductions of the surrounding provinces, are much larger than global-scale impacts.**

- 4) Considering the current modeling application over East Asia, the vertical 14 layers from surface to 10 km is too rough. First, the first layer is approximately 50m, but it will be usual to set 20-30m. The current model configuration is doubled thickness on first layer, and the representativeness as surface layer is ambiguous. Second, the upper model height is only 10 km. In my best knowledge, CMAQ does not support the top boundary condition. Therefore, this modeling system might have some problem to the treatment of stratospheric O<sub>3</sub>, and subsequently, to the model performance on surface level. The statistic analysis for O<sub>3</sub> (Table S2) seems to be out of range compared to the suggested model performance (Emery et al. 2017). Furthermore, this reproducibility for O<sub>3</sub> might lead to inaccuracy of other air pollutants. Reference) Emery et al. (2017, JA&WMA)

<https://www.tandfonline.com/doi/full/10.1080/10962247.2016.1265027>

**Response:** In previous manuscript, 14 layers refer to the vertical resolution of CMAQ model, while for WRF was 23 layers. We rewrite this part in Sect.2.4.1 and add some detailed information as follows. Similar vertical configurations for WRF and CMAQ model were applied in a lot of reported studies, and exhibited good agreement of PM<sub>2.5</sub> simulation with observations (Wang et al., 2010; Xing et al., 2011; Wang et al., 2012; Zhao et al., 2013; Zheng et al., 2015; Cai et al., 2017; Zheng et al., 2017; Campbell et al., 2018).

**Added/rewritten part in Sect.2.4.1:** The vertical resolution was designed as 23 sigma levels from surface to tropopause (about 100mb) for WRF simulation (with 10 layers below 3-km), while collapsed into 14 CTM layers by Meteorology-Chemistry Interface Processor (MCIP). The 14 sigma levels for CMAQ model vertical resolution were 1.000, 0.995, 0.988, 0.980, 0.970, 0.956, 0.938, 0.893, 0.839, 0.777, 0.702, 0.582, 0.400, 0.200 and 0.000.

**As this comment pointed out, O<sub>3</sub> was underestimated in this WRF-CMAQ model system, with the range of (-8.3%) - (-22.6%). The rough vertical layers, the underestimation of nature source emissions, the defect of upper boundary simulation in regional model and the uncertainties of VOCs emission inventories might all lead this underestimation. Given that the PM<sub>2.5</sub> concentration is the most important target in APPCAP, as well as the major pollutants this research focused on, the uncertainty introduced by O<sub>3</sub> simulation is relatively small, but further studies and efforts should made to improve the O<sub>3</sub> simulation. We also add a discussion in Sect.3.5.3 for this uncertainty.**

- 5) The emission inventory for Beijing is not taken from MEIC, but there is no reference and needs relevant information here. What was the difference between two inventories? Did the authors have specific reason to replace the emissions only for Beijing instead of MEIC?

**Response:** the emission inventory for Beijing (BJ-EI) in this study is from the Beijing Municipal Environmental Monitoring Center (BMEMC). We add a detailed description of BJ-EI in Sect.2.2; compare the major difference on

activity rate and spatial distribution of BJ-EI and MEIC in SI, Table S1; and explained why we replace the BJ-EI of MEIC in Sect.2.2 as follows. Mainly because the spatial distribution and emission source allocation of BJ-EI were more accurate than those of the MEIC model, which can significantly improve the air quality modelling, especially when modelling with finer resolutions (Zheng et al., 2017). Meanwhile, more detailed and objective activity rate, technology distribution and removal efficacy data at the county-level were collected from BJ-EI, which can largely reduce the uncertainty in estimating the emission reductions of each local control policy. However, similar local emission inventory as BJ-EI in other regions are not available, thus only BJ-EI can be replaced in MEIC.

**Added/rewritten part in Sect.2.2:** In this study, the anthropogenic emission inventory of Beijing was provided by the Beijing Municipal Environmental Monitoring Center (BMEMC). Based on the bottom-up method, the BMEMC developed a high-resolution emission inventory for Beijing (BJ-EI) of 2013 and 2017. BJ-EI basically had the same source classification as the MEIC model (described in Sect.2.3), however, the investigation and calculation process of BJ-EI were conducted at the county level, which of MEIC were conducted at the provincial level. The power, heating, industry (such as cement, iron, steel, chemical industry, manufacturing industry) and most solvent use (such as vehicle paint, ink, paint and coating) sectors were treated as point sources, with a higher accuracy of emission facility locations. In addition, fugitive dust emissions, including bare soil dust, road dust and construction dust, were added in BJ-EI, which were missing in MEIC model because of the lack of activity rates data. More detailed comparisons of BJ-EI and MEIC model can be found in SI, Table S1. Therefore, the spatial distribution and emission source allocation of BJ-EI were more accurate than those of the MEIC model, which can significantly improve the air quality modelling, especially when modelling with finer resolutions (Zheng et al., 2017). Meanwhile, more detailed and objective activity rate, technology distribution and removal efficacy data at the county-level were collected from BJ-EI, which can largely reduce the uncertainty in estimating the emission reductions of each local control policy.

- 6) What was the biomass burning inventory used in this study? I did not find the description.

**Response:** The open biomass burning inventory was not included in this study. On the one hand, the yearly emission changes are important in this study, however, current open biomass burning products (such as the Global Fire Emissions Database (van et al., 2017; <http://www.globalfiredata.org/>), the emission inventory of crop burning in China (Huang et al., 2012)) could not provide year-to-year emission variations, and the open biomass burning emission inventory for recent years was not available. On the other hand, although the lack of open biomass burning might introduce some uncertainties for the research, given that we mainly focused on the attribution of anthropogenic emission changes and meteorology impacts, the uncertainty introduced by this lack might be relatively small. We also add a discussion in Sect.3.5.3 for this uncertainty.

- 7) Section 2.4.2: In Table S2, I can only find the statistic for the year of 2017. Why did other years not shown? This section should be clearly separated into the description and discussion. Most of this section should be moved to subsection 3.2 or 3.1.

**Response:** please refer to the response for Specific Comments (10). We add the consecutive monthly descriptive statistic of PM<sub>2.5</sub> simulation during 2013-2017 in TableS3 (a), PM<sub>2.5</sub> compositions validation in Table S4 and other pollutants validations in Table S3 (b). We also add the analysis of these descriptive statistics in Sect.2.4.2.

- 8) Table 2: Does the parenthesis on rightmost column indicate observation? It should be clearly described.

**Response:** thanks for the kind remind. The parenthesis on rightmost column indicate the annual mean observational PM<sub>2.5</sub> concentrations of 2013, 2016 and 2017.

9) Typo: Section 3.4.2 should be 3.4.3

**Thanks for the kind remind; and the error is corrected in the new version.**

**References: (the references in bold texts are listed as follows, while the references in the added/written parts can be found in the manuscript)**

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