

Response to Reviewer I

General Comments:

- This manuscript studied the impact of meteorological changes and emission reductions on PM_{2.5} pollution in the Pearl River Delta (PRD) during the cold seasons from 2015 to 2017. The authors aimed to explain why PM_{2.5} levels in the PRD remained high despite of significant emission reductions in PRD and its upwind regions in East China. By applying the regional models, they found that transport contributions to PM_{2.5} levels rose from 70% in 2015 to 78% in 2017, while local emissions declined. And they concluded that the meteorology change was the dominant driver of the multiannual variations of PM_{2.5} during the studied period. And the meteorological change was likely driven by large scale climate variability, namely the transition from a strong El Niño in 2015 to a weak/moderate La Niña in 2017. This study also pointed out that the meteorological impact should be taken into consideration when the emission control policies were assessed.
- The manuscript is well organized and written clearly, the description is precise, and the discussion is fruitful. I recommend publishing it after minor revision. Below are my comments referring to lines (L), equations (Eqs.) and figures (Fig.).

Response:

We appreciate the valuable comments and suggestions. Our responses to specific comments are provided in blue and the corresponding revisions are highlighted in red. Please note that the line numbers (indicated by “Lx”) are those in the revised manuscript with the author’s changes.

Specific comments:

L160: spinning -> spanning

Response:

Thank you for noticing this spelling mistake. We corrected it as suggested.

L149: (1) Maybe it is better to specify the names of nine cities here when this concept is first mentioned.
(2) It seems that not all the nine cities are shown in Fig. 1.

Response:

(1) We provided the names of nine cities in L163-164 of the original manuscript. Since the concept of “nine cities” is firstly mentioned here, we moved the above information to L150-151:

... this study used PM_{2.5} monitoring data in the nine cities of the PRD (including Guangzhou, Shenzhen, Jiangmen, Zhuhai, Foshan, Dongguan, Zhaoqing, Huizhou and Zhongshan; Fig. 1), ...

(2) The names of cities have been added in the new Fig. 1:

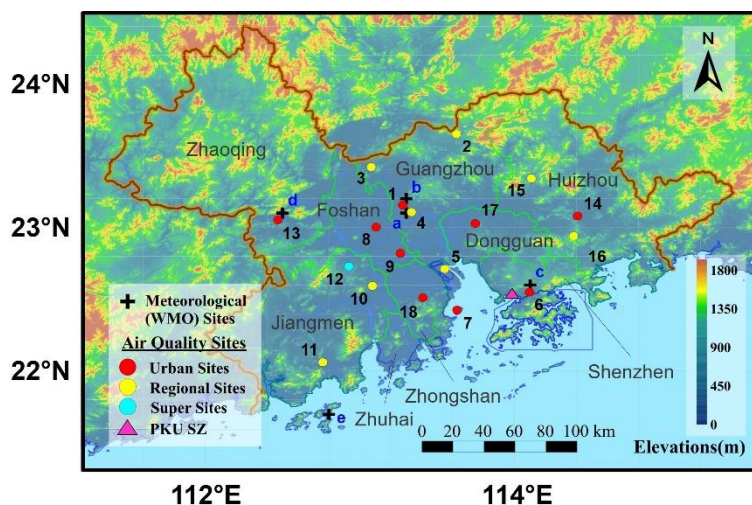


Fig. 1: (1) The x tick labels and y tick labels need to show the unit, e.g., 112 °E. (2) The orange line is not introduced in the caption.

Response:

(1) The axis labels have been revised, as suggested, in the new Fig. 1.

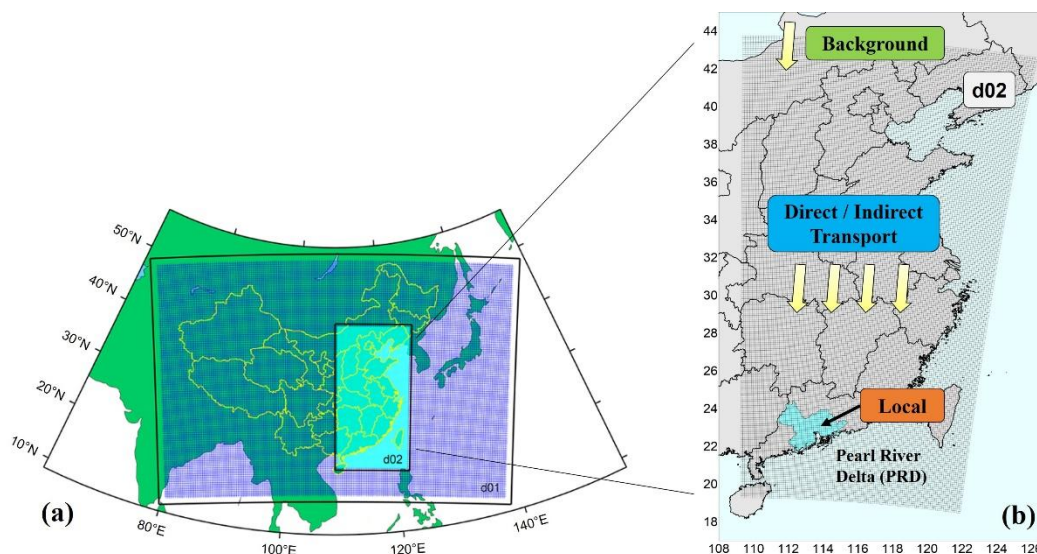
(2) We added the information of the orange line in the caption, in L161-162:

... The orange line indicates the boundary of the PRD region.

Fig. 2: (1) The latitude and longitude labels are too small. (2) "The black boxes are the simulation domains for WRF, while the nested areas indicate the simulation domains for CMAQ.": Does it mean that d01, d02, and the domain larger than d01 are all simulated by WRF? And CMAQ only simulates d01 and d02? Please specify it clearly.

Response:

(1) The axis labels have been revised in the new Fig. 2:



(2) Both WRF and CMAQ simulations are performed within d01 and d02, as introduced in the main text. However, the WRF domains are slightly bigger than the corresponding CMAQ domains, and that's why we used black boxes and nested areas to separately indicate the domains of two models. To avoid confusion, we've revised the sentence, as in L201-202:

The black boxes indicate the WRF simulation domains, which are slightly larger than the corresponding CMAQ domains, as represented by nested areas. Both WRF and CMAQ are applied to d01 and d02.

L190: Oct and Dec are selected in the simulations, but in L159, the cold season is defined as the period from Oct to Jan. Will this affect the simulation results?

Response:

We agree that focusing on two months in the simulations could potentially introduce some differences compared to the results for the full four-month cold season. However, the overall impact is likely minor.

PM_{2.5} transport to the PRD region during Oct-Jan is mainly driven by the East Asian winter monsoon, which maintains stable northeasterly wind patterns throughout this period (Ren et al., 2022). Although its intensity varies in different months, the general transport pathways and source contributions of PM_{2.5} in adjacent cold-season months remain similar.

We selected Oct. and Dec. to represent typical autumn and winter conditions, respectively, and used the combined results of these two representative months to approximate the overall cold-season state. The large amount of simulation hours (~1500 h per cold season) ensures statistically sufficient sampling to cover the cold-season variability in our analysis. More importantly, it restricts the simulation period, allowing for more efficient use of computational time and storage while still enabling robust investigation of PM_{2.5} sources and their inter-annual variations.

Eqs. 5 - 10: Why the contribution of $S_{Emis_O,15/16}$ is not calculated by $C_{L15O16M15} - C_{Base15}$? And similar questions for $S_{Meteo,15/16}$, $S_{Emis_O,16/17}$, and $S_{Emis_L,16/17}$?

Response:

Thank you for this comment. To clarify, in theory, the contribution of $S_{Emis_O,15/16}$ could be estimated by directly comparing the simulation results of the L15O16M15 and Base15 scenarios. However, we adopted the current design of simulation scenarios because it captures the accumulated effects of changes in local emissions, outer emissions and meteorology, while ensuring that the sum of the three individual contributions between two adjacent years equals the net change in concentrations or source contributions. This approach of designing simulation scenarios has been widely applied in similar studies (Jiang et al., 2022).

Fig. 4: The x tick labels and y tick labels need to show the unit, e.g., 110 °E.

Response:

The axis labels have been revised in the new Fig. 4:

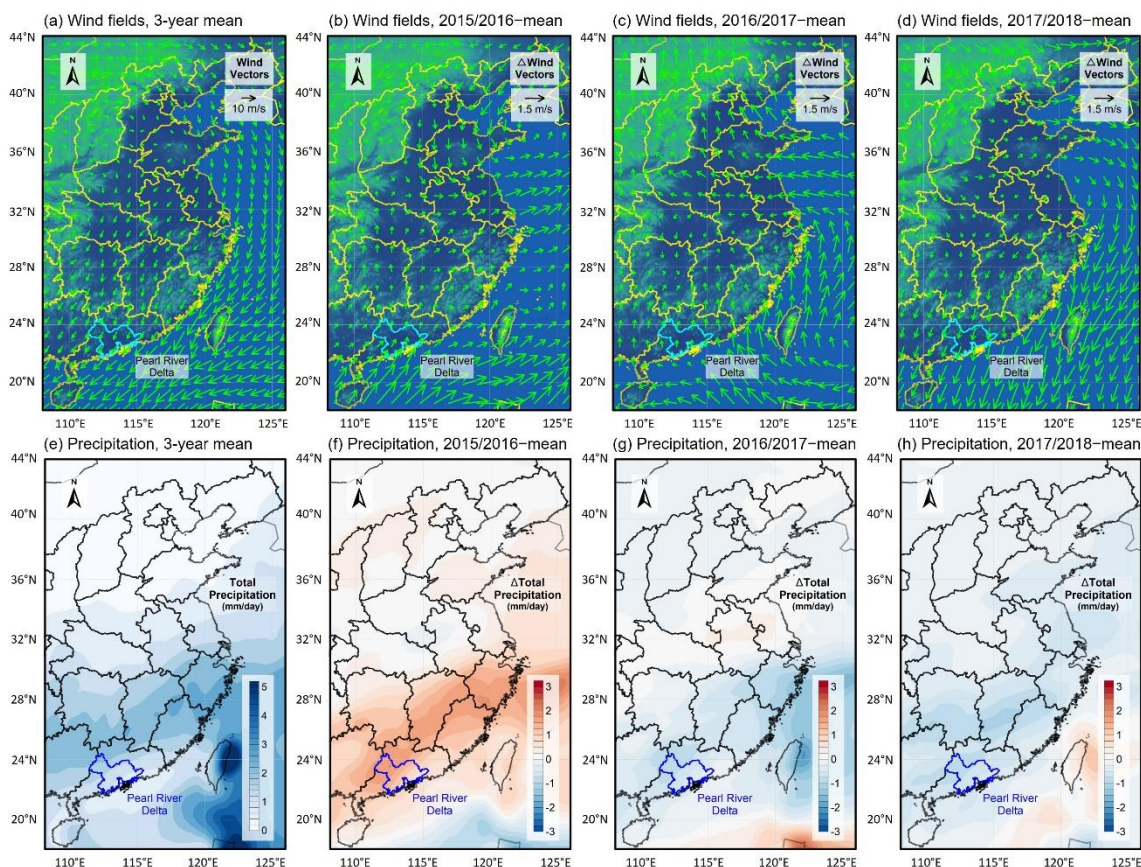
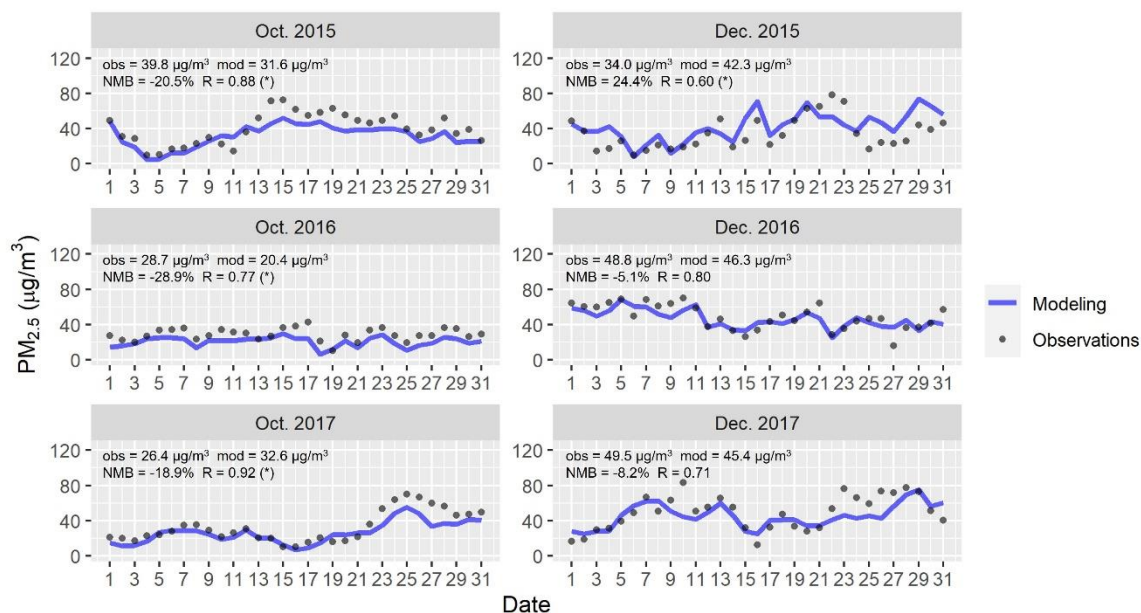


Fig. 5: The legend of "Observations" is point instead of point+line.

Response:

The legend of "Observations" has been corrected in the new Fig. 5:



L412: What does 'population-weighted mean' represent?

Response:

The population-weighted mean concentration of pollutants is a widely used metric in air quality assessment studies, especially those focusing on human exposure (e.g., Li et al., 2017). In comparison to the normally used area-weighted concentration, this metric assigns greater weight to regions with higher populations, thereby providing a more representative estimate of the health effect of air pollutants. We have added more details on this metric in the Method section, in L253-259:

The simulated population-weighted pollutant concentration is used for further source apportionment calculation and analysis. As it better indicates the effect of air pollutants on human health, this metric is widely used in air quality assessment studies (e.g., Li et al., 2017c). The population-weighted concentration ($f_{pop-weighted}$) is calculated as follows:

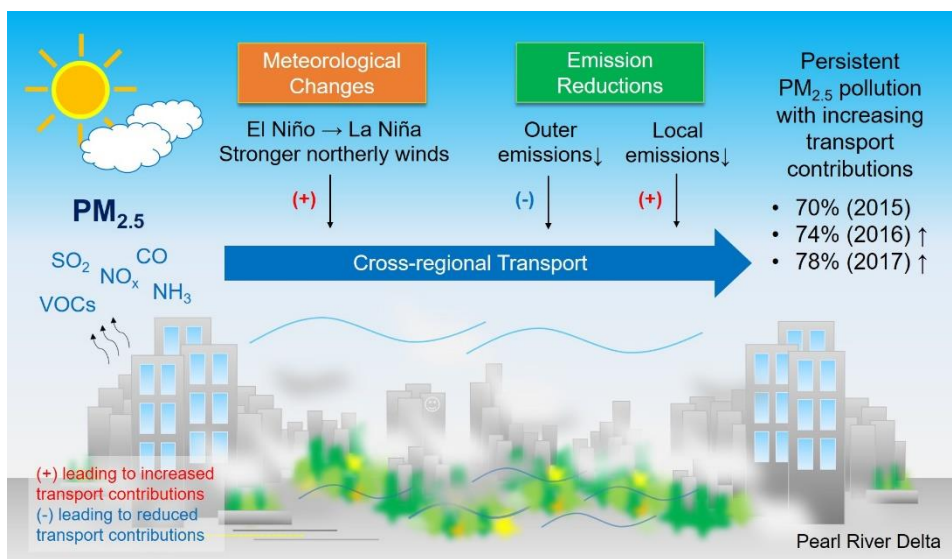
$$f_{pop-weighted} = \frac{\sum_{i=1}^N f_i p_i A_i}{\sum_{i=1}^N p_i A_i} \quad (1)$$

where f_i is the simulated pollutant concentration in the grid cell i ; p_i is the population density within the grid cell i ; A_i indicates the area of the administrative PRD region within the grid cell i ; N is the total number of grid cells within the simulation domain. Gridded population density data for the year 2015 were obtained from the GPWv4 dataset (last access: 14 September 2017; Doxsey-Whitfield et al., 2015) and applied in the above calculation.

Fig. Graphic Abstract: Why the decrease of local emissions is marked with "Enhanced"?

Response:

It means the decrease of local emissions contributed to the increase in transport contributions, or enhanced the effect of cross-regional transport. To make it clear, we've revised the Graphic Abstract, as follows:



References

- Doxsey-Whitfield, E., MacManus, K., Adamo, S. B., Pistolesi, L., Squires, J., Borkovska, O., and Baptista, S. R.: Taking advantage of the improved availability of census data: a first look at the gridded population of the world, version 4, *Papers in Applied Geography*, 1, 226–234, <https://doi.org/10.1080/23754931.2015.1014272>, 2015.
- Jiang, Y., Wang, S., Jia, X., Zhao, B., Li, S., Chang, X., Zhang, S., and Dong, Z.: Ambient fine particulate matter and ozone pollution in China: synergy in anthropogenic emissions and atmospheric processes, *Environ. Res. Lett.*, 17(12), <https://doi.org/10.1088/1748-9326/aca16a>, 2022.
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Response to Reviewer II

General Comments:

This manuscript presents a valuable investigation into the complex interplay between emission reductions and meteorological variability on PM_{2.5} pollution in the Pearl River Delta from 2015 to 2017. The study is well-designed, employing the WRF/CMAQ model with a comprehensive set of sensitivity analyses to deconstruct the drivers of the observed persistent pollution. The paper is clearly written, and the conclusions are well-supported by the analysis. I enjoyed reading this manuscript. This paper is suitable for publication after some minor revisions.

Response:

We appreciate the valuable comments and suggestions. Our responses to specific comments are provided in blue and the corresponding revisions are highlighted in red. Please note that the line numbers (indicated by “Lx”) are those in the revised manuscript with the author’s changes.

Specific comments:

1) A minor suggestion for the title. While "Persisted" is accurate, a word like "Anomalous" or "Resurgent" might better capture the unexpected increasement of PM_{2.5} during 2015-2017. This is just a suggestion for the authors to consider.

Response:

Thanks for this helpful suggestion. By using the word “persisted” (or “persistent”), we intended to stress that PM_{2.5} pollution showed no significant improvement over the three-year period. However, we agree that highlighting the unexpected nature of this persistence is also important to show the significance of our findings. Therefore, we revise the title into:

Unexpectedly persistent PM_{2.5} pollution in the Pearl River Delta, South China, in the 2015-2017 cold seasons: The dominant role of meteorological changes during the El Niño-to-La Niña transition over emission reduction

2) In Section 2.3 (line 213), the manuscript introduces the Factor Separation Method (FSM). Could the authors first confirm if this method is fully described by Equations 1-4? Second, a brief discussion on the advantages of FSM over the more traditional Brute Force Method (BFM), such as its ability to quantify the interactive effects between different emission regions, would be beneficial for readers less familiar with the technique.

Response:

Thank you for this comment. We’ve rechecked Equations 1-4 and confirm that the FSM is fully described. In the original manuscript, we briefly introduced FSM, in L220-222:

This approach enables a detailed assessment of how much local and external emissions contribute to PM_{2.5} pollution while also identifying their interactive effects, thus it has been applied in many previous studies (Chen et al., 2014; Uranishi et al., 2018; Qu et al., 2021b; Sun et al., 2022; Xu et al., 2023).

To improve the text, we have now added further discussion on the advantages of FSM, in L240-243:

Currently, these contributions can only be identified by the FSM approach, whereas other source apportionment methods (e.g., top-down or bottom-up Brute Force Method and the tagging method; Clappier et al., 2017) typically classify or separate them into either local or external contributions, highlighting the advantage of this method for our study.

3) Regarding the background contribution (F_{bg}), the definition could be more explicit. Please clarify in the text that this term represents all influences from outside the d02 simulation domain, as determined by the chemical boundary conditions used in the model.

Response:

We appreciate the reviewer's comment. The definition of the background contribution has now been made more explicit, in L232-233:

Background contribution (F_{bg}): Contribution from sources outside the d02, estimated as the contributions of chemical boundary conditions used in the d02 simulations.

4) Could the authors provide a more explicit discussion on the specific meteorological drivers for the changes in sulfate and nitrate? For instance, was the significant change in nitrate primarily driven by shifts in temperature and humidity (impacting chemical formation) or by changes in wind patterns (impacting transport)? Disentangling these influences would provide a clearer mechanistic understanding.

Response:

We thank the reviewer for this suggestion. The responses of sulfate and nitrate to meteorological changes are discussed in the Sections 4.3 and 4.4. We found that sulfate is not sensitive to meteorological changes, and explanations were added in L518-523:

Unlike $PM_{2.5}$, which was strongly affected by meteorological changes, pSO_4 exhibited a relatively limited response to them. This may be attributed to the high proportion of pSO_4 originating from cross-regional transport (80-90%) compared to local contribution (10-20%). The transport patterns remained generally consistent (Fig. 4a), resulting in insignificant changes in pSO_4 contributed by transport. Although locally produced pSO_4 can be influenced by local meteorological conditions, its small contribution makes the responses of the overall pSO_4 level to local meteorological changes less notable.

Unlike sulfate, nitrate was notably influenced by meteorological changes, as stated in L525-528:

Also, these changes were largely driven by varying meteorological conditions, which influenced all major pNO_3 source contributions, resulting in their declines during 2015-2016 and increases during 2016-2017. More analyses of the meteorological influences on pNO_3 will be presented in the next section.

The reasons behind reduced nitrate are provided in L601-608:

The contributions of other processes to $PM_{2.5}$ exhibit diversified variations, indicating complex responses of these processes to meteorological and emission changes. Overall, meteorological changes played a more important role in driving these variations. They resulted in a reduction in the contribution of aerosol process (stronger negative contributions during the daytime and weaker positive contributions at night),

which is likely associated with the enhanced partitioning of pNO_3 into gas-phase HNO_3 . $\varepsilon(\text{pNO}_3)$, the proportion of pNO_3 in the sum of pNO_3 and gas-phase HNO_3 , decreased significantly across the three cold seasons (from 70.6% in 2015 to 67.7% in 2016 and 61.7% in 2017; Table S4), suggesting a greater tendency for pNO_3 to shift into the gas phase. It can be primarily attributed to meteorological changes over the three years, namely, higher temperature in 2016 and reduced RH in 2017 (Table 3).

To clearly summarize the responses of sulfate and nitrate to meteorological changes, we revised the conclusion part, in L650-654:

... Emission reduction, particularly outside the PRD, led to consistent decreases in pSO_4 concentrations in the three cold seasons, whereas the influence of meteorological changes was overall limited due to the high transport (and low local) contribution to pSO_4 . In contrast, the three-year changes in pNO_3 concentrations were largely controlled by meteorological variations, likely associated with the varied partitioning between particle-phase pNO_3 and gas-phase HNO_3 under changes in local temperature and humidity.

5) In Sections 4.3 and 4.4, the text describes complex trends from the source apportionment and budget analyses. While the figures and tables are comprehensive, the narrative would be much easier to follow if key quantitative values were integrated directly into the descriptive text. This would reduce the need for the reader to constantly switch between the text, figures, and tables.

Response:

We thank the reviewer for the helpful suggestions. To make the Sect 4.3 and 4.4 more reader-friendly, we've added representative quantitative values in the texts, e.g. in L494-496:

As expected, reductions in local and outer emissions lowered local and direct transport contributions to $\text{PM}_{2.5}$, respectively, leading to overall decreases in $\text{PM}_{2.5}$ concentrations during both 2015-2016 (by 1.2 and $4.1 \mu\text{g m}^{-3}$, respectively) and 2016-2017 (by 1.7 and $1.0 \mu\text{g m}^{-3}$, respectively).

In L516-517:

pSO_4 : Its concentration showed a consistent decline throughout the study period, dropping from $9.6 \mu\text{g m}^{-3}$ in 2015 to $7.3 \mu\text{g m}^{-3}$ in 2016 and $6.3 \mu\text{g m}^{-3}$ in 2017.

In L524-525:

pNO_3 : pNO_3 concentrations followed a similar change pattern as $\text{PM}_{2.5}$, decreasing from $8.2 \mu\text{g m}^{-3}$ to $5.3 \mu\text{g m}^{-3}$ during the first two cold seasons but rising to $6.6 \mu\text{g m}^{-3}$ in 2017.

In L541-543:

pNH_4 concentrations declined from $5.8 \mu\text{g m}^{-3}$ in 2015 to $4.1 \mu\text{g m}^{-3}$ in 2016 but remained stable at $4.0 \mu\text{g m}^{-3}$ in 2017. The influences of meteorological and emission changes on pNH_4 were comparable ($-0.7 \mu\text{g m}^{-3}$ vs. $-0.9 \mu\text{g m}^{-3}$ during 2015-2016; $+0.4 \mu\text{g m}^{-3}$ vs. $-0.5 \mu\text{g m}^{-3}$ during 2016-2017).

In L583-587:

Overall, at different times of the day, both the total $\text{PM}_{2.5}$ flux transported into and out of the PRD (hereafter referred to as $\text{PM}_{2.5}$ influx and outflux, respectively) increased across the three cold seasons when emissions were fixed at the 2016 level. For instance, the morning $\text{PM}_{2.5}$ influx rose from 225.4 t h^{-1}

in 2015 and 228.5 t h⁻¹ in 2016 to 298.4 t h⁻¹ in 2017, while the afternoon PM_{2.5} outflux increased from -292.3 t h⁻¹ in 2015 to -302.2 and -314.2 t h⁻¹ in 2016 and 2017, respectively.

In L593-596:

However, the morning influxes and afternoon outfluxes of ABLex-H did not show a continuous decrease over the three years. Instead, their values were relatively higher in both the 2015 and 2017 cold seasons (151.7 and 132.8 t h⁻¹ for morning influxes, and -264.9 and -220.5 t h⁻¹ for afternoon outfluxes) compared with those in 2016 (112.0 and -212.6 t h⁻¹).

And in L605-607:

$\epsilon(\text{pNO}_3)$, the proportion of pNO₃ in the sum of pNO₃ and gas-phase HNO₃, decreased significantly across the three cold seasons (from 70.6% in 2015 to 67.7% in 2016 and 61.7% in 2017; Table S4), suggesting a greater tendency for pNO₃ to shift into the gas phase.

These additional notes will help readers grasp the key quantitative results without referring constantly to the figures and tables.

6) The conclusion that unfavorable meteorology can mask emission control benefits is powerful and has significant policy implications. This point could be further nuanced by adding a brief discussion in the final section about the differing timescales of these effects. While meteorological variability can clearly dominate inter-annual changes, it is crucial to emphasize that sustained, long-term emission reductions remain the fundamental and most effective strategy for improving air quality.

Response:

We appreciate this insightful suggestion and fully agree that meteorological effects and emission control act on PM_{2.5} pollution variations on different timescales. While our results show that meteorological changes can temporarily offset the effects of emission control, it is important to mention that emission reductions should remain the primary strategy for long-term air quality improvement. In response to this comment, the following text has been added in the end of Conclusions, in L684-686:

Although meteorological variability can drive multi-annual changes in regional PM_{2.5} pollution, sustained emission reductions remain the most fundamental and effective means to achieve long-term air quality improvement.

References

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