Reviewer 2

This manuscript investigates drivers of PM2.5 and PM10 changes in the Beijing–Tianjin–Hebei (BTH) and Yangtze River Delta (YRD) regions between 2015–2020 by combining national monitoring data, GEOS-FP meteorological fields, the CEDS emissions inventory, and a LightGBM machine-learning model. This manuscript documents significant declines in PM concentrations and attributes most of the reductions to anthropogenic emission decreases, while identifying specific meteorological variables and pollutant co-variations that modulate PM variability. Generally, I think the topic of this study is within the scope of ACP journal. The dataset and method applied here are reasonable. This manuscript is also well written, structured and analyzed convincingly. I recommend that this manuscript can be published in ACP after revisions.

Response: We thank this reviewer for his comments. We will respond to his comments point by point as shown below.

Major concerns:

1. Please explain the meaning of "co-emissions-chemical transformation-meteorological synergy". This term is not explained in the paper but appears frequently in the abstract and main text.

Response: Thank you for raising this point. In the original submission, the phrase *"co-emission–chemical transformation–meteorological synergy"* appeared in the abstract and discussion, but we agree that the terminology was not clearly defined and could lead to conceptual ambiguity.In the revised manuscript, we have removed this expression and replaced it with a more precise and descriptive wording—*"emission–chemical transformation–meteorological coupling"*. This updated phrasing is used only in the Discussion section and is directly supported by the physical mechanisms quantified in our analysis, including (1) the covariation of $CO/NO_2/SO_2$ with $PM_{2.5}$ due to shared combustion sources, (2) the shift from sulfate- to nitrate-dominated chemistry following nationwide desulfurization, and (3) the strong temperature dependence of NO_x-driven secondary formation.By using this clearer terminology and linking it explicitly to the SHAP-based mechanistic evidence, the revised manuscript avoids the ambiguity of the previous expression while retaining an accurate interpretation of the processes described.

2. This article reveals the impact of anthropogenic emissions on PM2.5 and PM10. I am very curious whether this effect is consistent with the trends in anthropogenic emission inventories. It would be very meaningful if this method could be used to verify emission inventories.

Response: Thank you for this important comment. To address your question, we examined whether the SHAP-derived emission contributions are temporally consistent with the anthropogenic emission inventories used as model inputs. The new Fig. 6 (in the revised manuscript) compares the monthly evolution of NO, SO_2, and BC emission inventories with their corresponding SHAP contributions. For all three species, the SHAP time series closely follow the temporal patterns of the inventories, with strong positive correlations (R = 0.89–0.95). This consistency indicates that the model attribution is sensitive to and reflects the real temporal variability of the emission inputs, providing confidence that the LightGBM–SHAP framework is capturing physically meaningful emission-driven signals.

The revised manuscript includes these results (Fig. 6; See lines 405–418).

3. The introduction to the methods of calculating feature importance is missing. Please introduce the method for calculating feature importance.

Response: Thank you for this comment. We have added a clear introduction to the method used for calculating feature importance in the revised manuscript. Specifically, we now describe the SHAP (Shapley Additive explanations) framework and how it quantifies the marginal contribution of each predictor to LightGBM model outputs. This new methodological description has been incorporated into Section 3.2.3 (lines 265–280).

4. For SO2 and NO2 correlations, please elaborate on how these relate to specific primary emission control measures (e.g., desulfurization, denitrification) and shifts in secondary aerosol formation pathways, to better substantiate the proposed "synergy" mechanism.

Response: Thank you for this excellent suggestion. In the revised manuscript, we have added a clear explanation linking the SO_2/NO_2 correlations to real emission-control measures and associated chemical regime transitions. Specifically, the Discussion (Section 5) now clarifies that :(1) Weaker SO_2 correlations reflect the strong impact of nationwide coal desulfurization, which has greatly reduced sulfate production and shifted secondary inorganic aerosol chemistry toward nitrate dominance. (2) Intermediate NO_2 correlations indicate that although denitrification has progressed, traffic and industrial NO_x emissions remain important precursors, consistent with the increasing dominance of nitrate in recent years. These additions strengthen the interpretation of the SO_2 and NO_2 correlation patterns using updated terminology, consistent with the revisions made elsewhere in the manuscript (see lines 4490–463).

5. The strong PM2.5–CO correlations and their source attribution implications are a central result of this study. However, recent multi-platform observation and top-down constrained studies have revealed similar mechanisms linking CO, NO_x, and carbonaceous aerosols to combustion-related PM2.5 sources. For example, Wang et al. (2025, npj Climate and Atmospheric Science), Wang et al. (2021, Earth's Future), and Tiwari et al. (2025, Communications Earth & Environment) provide global and regional evidence that such emission linkages are also major contributors to CO2 and black carbon emissions, consistent with the "co-emission–chemical transformation–meteorological synergy" framework. Citing these works would contextualize your findings within the broader emission research landscape and strengthen the scientific relevance of your results.

Response: Thank you for recommending these important papers. We have now: (1) Cited all three works (Wang et al., 2025; Wang et al., 2021; Tiwari et al., 2025) in the Discussion. (2) Related our $PM_{2.5}$–CO findings to the broader evidence presented in these studies regarding the coupling among combustion emissions, precursor gases, and secondary aerosol formation. (3) Positioned our SHAP-derived $PM_{2.5}$–CO results as consistent with—and extending beyond—these earlier analyses by examining a more recent period (2015–2022), including both $PM_{2.5}$ and PM_{10}, and incorporating temperature–NO_x interactions that further constrain secondary inorganic aerosol production. We also note that the terminology used in the initial submission has been replaced with more neutral wording throughout the revised manuscript (see lines 449–489).

Minor concerns:

1. There are many places in this article where there are no spaces such as P8, Lines 200, and P9, Lines 205. Please check and improve it.

Response: Thank you for pointing this out. We have carefully checked the entire manuscript and corrected all missing-space issues.

2. The color bars in many images should be adjusted to correspond to positive and negative values. For example, the range in Figure 1 is -9 to 3, which should be changed to -9 to 9. Please check and improve it.

Response: Thank you for this helpful comment. We have thoroughly checked all figures containing color bars and revised their ranges where appropriate. For plots that include both positive and negative values, the color bars have been adjusted to symmetric ranges (e.g., −15 to +15 in Figure 1) to improve visual consistency. For figures whose data are strictly non-negative (e.g., correlation coefficients ranging from 0 to 1) or strictly non-positive (e.g., uniformly negative trends), symmetric color bars are not applicable because the underlying quantities do not span both signs. In these cases, the original directional ranges have been retained to accurately reflect the data distribution. All figures have now been reviewed and updated accordingly.

3. The image sizes in Figure 2 should be kept consistent.

Response: Thank you for your comment. The panel sizes in Figure 2 have been checked and adjusted to ensure consistent image dimensions across all subplots.

4. P20, Lines 435, Please give the full name of PMF, CMB.

Response: Thank you for your comment. In the revised manuscript, the references to PMF and CMB were removed during content restructuring, as these terms were no longer needed in the final version. Therefore, their full names are not required in the current manuscript.

5. Please check this manuscript for grammatical errors.

Response: Thank you for your comment. We have carefully checked the entire manuscript for grammatical and typographical errors and corrected all issues identified.

6. In the reference list, some entries have inconsistent journal name abbreviations (e.g.,7. 8. “Atmospheric Chem. Phys.” vs. “Atmos. Chem. Phys.”) and DOI formatting — unify them according to ACP style.

Response: Thank you for these helpful suggestions. We have reviewed and standardized all journal abbreviations and DOI formats in the reference list to ensure consistency with ACP style.

7. Occasional double spaces or missing spaces between words and numbers (e.g., “to2020” in

p. 5 line 117 should be “to 2020”).

Response: Thank you for these helpful suggestions. All occurrences of missing or double spaces (e.g., “to2020”) have been corrected throughout the manuscript.

8. The Zenodo DOI in line 452 is presented without a clickable hyperlink. For ACP style, this should be fully hyperlinked.

Response: Thank you for these helpful suggestions. The Zenodo DOI has been reformatted as a fully clickable hyperlink, consistent with ACP formatting requirements.