Response Letter

The revision has addressed some of the comments in the first round of review. I would like to encourage the authors to consider the following further comments before it can be accepted for publication.
Response: We thank the reviewer for the constructive comments. We respond to each point in detail below.

1) According to the description, for every city, pollutant and fold, there are at most 84 data records for the model training (namely, 7 years with 12 months each) of one single LightGBM model. Is this really enough? I am very concerned about overfitting. I refer to papers like https://acp.copernicus.org/articles/19/11303/2019/, where thousands of data records are used for training one model. Please provide evidences to support that overfitting is not a concern for your model training.
Response: We acknowledge the reviewer's concern regarding the limited number of monthly samples per city and the potential risk of overfitting. To address this, we adopted a strictly temporally separated leave-one-year-out (LOGO) cross-validation scheme, in which each test year is completely excluded from both model fitting and hyperparameter tuning. This design provides a conservative out-of-sample evaluation and avoids any within-year information leakage. To further assess whether the available sample size is sufficient, we performed learning-curve analyses for $PM_{2.5}$ and PM_{10} under the same LOGO framework (Supplementary Fig. S5, S6). The learning curves show stable convergence, with model performance improving rapidly at smaller training sizes and reaching a clear plateau beyond approximately 48–60 months. No systematic degradation or instability in validation performance is observed with increasing training size, indicating that overfitting is not a major concern despite the limited number of monthly samples.

2) Figure 2: Does it make sense to exclude certain cities with very low predictions power (e.g., cities with correlation < 0.7, so $R2<0.5$, meaning that less than 50% variability can be explained by the LightGBM model)?
Response: Regarding the reviewer's question on whether cities with lower predictive skill should be excluded, we emphasize that reduced performance in some cities does not indicate model instability or overfitting. As demonstrated by the learning-curve analyses (Fig.S5,S6), the modeling framework exhibits consistent generalization behavior across a wide range of training sample sizes. The variability in predictive skill, particularly for PM_{10}, is more plausibly attributed to stronger local heterogeneity and non-stationarity of coarse-particle processes rather than to deficiencies in the modeling approach. Excluding cities with lower predictive performance could introduce selection bias and would disproportionately emphasize more easily predictable conditions. For this reason, all cities are retained in the analysis to preserve a comprehensive and unbiased regional assessment.

3) PM10 contains a significant contribution from PM2.5. So to distinguish the drivers for fine and coarse particles, does it make more sense to (maybe additionally) examine

the coarse mode mass (PM2.5-10), and contrast the results vs. PM2.5? It appears to me that the current discussion still lacks strong distinction, e.g., Figure 3 still suggests very correlated feature importances for PM2.5 and PM10, possibly due to the inclusion of PM2.5 in PM10.

Response: We thank the reviewer for this insightful comment. We agree that PM_{10} inherently includes a substantial contribution from $PM_{2.5}$, which can lead to similar feature importance patterns for $PM_{2.5}$ and PM_{10}. To address this point, we have revised the manuscript to explicitly clarify the physical interpretation of PM_{10} at its first appearance in the SHAP-based analysis (Lines 366–381), explaining that the similarity in Fig. 3 primarily reflects shared driving processes associated with the fine particle fraction embedded in PM_{10}, rather than a limitation of the modeling framework. In addition, following the reviewer's suggestion, we conducted a supplementary analysis of the coarse-mode mass defined as $PM_{2.5\text{-}10}$ (PM_{10} minus $PM_{2.5}$) using the same leave-one-year-out LightGBM–SHAP framework. The resulting feature importance rankings exhibit systematic differences from those of PM_{10}, demonstrating that the model is capable of distinguishing size-dependent drivers when the fine particle contribution is explicitly removed. The corresponding results are provided in the Supplementary Material (Fig. S7). Finally, we note in the Conclusions that a more comprehensive size-resolved analysis will be pursued in future work to further investigate fine particle and coarse particle processes (Lines 552-556).

4) Line 363-366: why do humidity and temperature have stronger modulation roles on coarse particles than fine particles? I think many papers suggests the opposite (fine particles are more sensitive to these factors), and coarse particles (e.g., mainly dust over land) should have stronger dependences on wind speed and soil moisture? Overall, this statement appears not convincing without further discussion with existing literature results.

Response: We thank the reviewer for this helpful comment and for pointing out the apparent inconsistency with existing literature. We agree that fine particles ($PM_{2.5}$) are generally more sensitive to humidity and temperature, whereas coarse particles are often more closely associated with wind-driven emission and surface conditions. The reviewer's concern is related to the wording in Lines 363–366 of the original manuscript, which may have led to an overinterpretation of the role of humidity and temperature for coarse particles. In the revised manuscript, this part has been carefully clarified. We now explicitly state that PM_{10} is an integrated metric that contains a substantial fine particle fraction, and that the high SHAP importance of humidity- and temperature-related variables for PM_{10} should be interpreted in this context, rather than as evidence of a stronger sensitivity of coarse particles. In addition, as shown in the Supplementary Material (Fig. S7), we performed an additional analysis of $PM_{2.5\text{-}10}$ to provide a more direct diagnostic of coarse-mode variability. The resulting feature importance patterns differ systematically from those of PM_{10} and are more consistent with the reviewer's expectations for coarse particles. We believe that these revisions improve the clarity and consistency of the interpretation while remaining aligned with existing literature.

5) Figure 5: it seems a little hard for me to understand that the meteorology-driven changes (blue) are smaller for PM10 than for PM2.5. Why?

Response: We thank the reviewer for this question. Fig. 5 shows the net contribution of meteorological factors to interannual PM variability, rather than instantaneous sensitivity to individual meteorological processes. The smaller meteorology-driven changes for PM_{10} therefore do not imply a weaker meteorological influence on coarse particles. For $PM_{2.5}$, meteorological effects such as temperature, humidity, and boundary-layer conditions tend to influence secondary formation and hygroscopic growth in a relatively consistent direction at the interannual scale, allowing their impacts to accumulate into a clearer net contribution. In contrast, the meteorological influences on PM_{10} are often more process-dependent and can act in opposite directions (e.g., wind-enhanced resuspension versus enhanced dispersion and removal, or precipitation-related wet deposition), leading to partial cancellation when aggregated at annual timescales. As a result, although PM_{10} can be highly sensitive to meteorological conditions at shorter timescales, its net meteorology-driven contribution to interannual variability can appear smaller when compared to $PM_{2.5}$. We have clarified this interpretation in the revised manuscript to avoid potential confusion (Lines 411–429).

6) Also Figure 5: why is the meteorology-driven changes always positive? Is it because of the highly positive values in Figure 4 for certain cities? If so, does it make sense to add a distribution of numbers among cities in Figure 5?

Response: We thank the reviewer for raising this point. The consistently positive meteorology-driven contributions in Fig. 5 reflect the definition and aggregation level of the analysis, rather than uniformly positive meteorological effects across all cities. Specifically, Fig. 5 summarizes interannual meteorology-driven deviations relative to a long-term baseline and then averages these deviations across cities. As a result, the values represent net regional effects at the annual scale, rather than city-specific responses. At the city level, meteorological contributions exhibit substantial spatial heterogeneity, with both positive and negative values, as already shown in Fig. 4. When aggregated across cities, opposing meteorological influences (e.g., enhanced dispersion versus unfavorable stagnation conditions) partially offset each other, leaving a weakly positive net contribution in most years. This behavior is consistent with previous attribution studies that decompose interannual variability at regional scales. Because the city-level distributions and their variability are explicitly presented in Fig. 4, Fig. 5 is intended to focus on the regional-mean interannual evolution. We therefore did not add an additional distribution plot in Fig. 5 to avoid redundancy, but we have clarified this interpretation in the revised manuscript (Lines 411–429).

7) Figures 6 and 7: I do not understand the meaning of the light blue shadings?

Response: In the revised manuscript, the light-blue shaded area previously shown in Figs. 6–7 has been removed. In the original version, this shading was intended only as a visual aid to highlight the separation between the monthly mean inventory series and the corresponding SHAP-based temporal series for the same predictor. However, we recognize that such a shaded region may be misinterpreted as an uncertainty band or confidence interval. To avoid any potential confusion, we have eliminated the shaded area in the revised figures and now present only the two time series using separate y-axes. The figure captions have been updated accordingly.

8) Line 416-419: I can list many papers discussing high temperature and/or high moisture facilitate more PM, examples are below:

https://www.pnas.org/doi/abs/10.1073/pnas.2022179118
https://www.science.org/doi/full/10.1126/science.adq2840
https://www.sciencedirect.com/science/article/pii/S1352231013004743
https://pubs.acs.org/doi/full/10.1021/acsearthspacechem.2c00077
https://www.science.org/doi/full/10.1126/science.adg8204

While you suggest the opposite. Please provide more evidence with possible mechanisms to further support your argument.

Response: We thank the reviewer for pointing out this important issue and for providing relevant references. We fully agree that a substantial body of literature demonstrates that high temperature and humidity can facilitate particulate matter formation, particularly for $PM_{2.5}$, through processes such as enhanced secondary aerosol production and hygroscopic growth. In our study, however, the discussion in Lines 416–419 refers to meteorology-driven contributions evaluated at the interannual scale and in a relative sense, namely deviations from a long-term baseline after explicitly separating emission-driven trends. Under this attribution framework, the reported effects should be interpreted as net annual contributions rather than instantaneous or process-level sensitivities. It is also important to note that particulate pollution in China exhibits strong seasonal characteristics, with the highest PM concentrations typically occurring during winter. Consequently, when temperature is examined at the interannual scale, higher annual-mean temperatures may coincide with weaker wintertime stagnation and reduced heating-related emissions, which can result in an apparent inverse relationship between temperature and PM variability after emission trends are removed. This behavior reflects the seasonal structure of pollution and does not contradict established process-based findings. Similarly, humidity in this study is represented by regional-scale meteorological reanalysis data and evaluated at monthly to annual timescales. At these scales, humidity integrates multiple competing influences, including aerosol water uptake, precipitation-related removal, and circulation-driven transport, while local or microphysical humidity effects cannot be fully resolved. As a result, our analysis emphasizes the relative importance of humidity as a modulating factor in long-term variability rather than attempting to isolate individual process-level mechanisms. To avoid confusion, we have revised the manuscript to clarify that our findings do not contradict existing studies. Instead, they highlight that the sign and magnitude of meteorological effects on particulate matter can differ across temporal scales and attribution frameworks. We have also adjusted the wording in the revised text to better align with the established literature and to emphasize the scale-dependent nature of meteorological influences on particulate matter (Lines 438–447).

9) Section 5, Figure 8 and Figure 9: I do not find a strong connection between these correlation analysis with "physically interpretation of the SHAP analysis results". There is no systematic pattern of the "SHAP heat map" in Figure 9. I suggest to completely delete this part.

Response: We thank the reviewer for this thoughtful comment. We agree that the correlation analysis and the SHAP heat maps presented in Section 5 are not intended to provide a direct or standalone physical interpretation of particulate matter processes. Instead, this section is included to offer a supplementary perspective on whether the SHAP-derived temporal contributions exhibit statistically meaningful associations with corresponding emission and meteorological indicators. We acknowledge that the SHAP heat map in Fig. 9 does not display a single systematic pattern across all variables and regions. This behavior reflects the heterogeneous and non-stationary nature of emission–meteorology interactions across cities and years, rather than a limitation of the SHAP framework itself. To avoid overinterpretation, we have revised the manuscript to clearly state the limited and contextual role of Section 5, and to emphasize that the main physical interpretation relies on the SHAP feature importance rankings and contribution analyses presented in earlier sections. With this clarification, we retain Section 5 as a complementary analysis that provides additional context for the attribution results, while deliberately avoiding strong mechanistic claims based solely on correlation or heat-map patterns. We believe that this revision improves clarity without affecting the main conclusions of the study (Lines 472-509).