

1 **Quantifying the driving factors of particulate matter variabilities in the Beijing-Tianjin-Hebei**  
2 **and Yangtze River Delta regions from 2015 to 2022 by machine learning approach**

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16  
17 **Abstract.** Accurately quantifying the relative roles of anthropogenic emissions and meteorological  
18 conditions is essential for understanding long term changes in particulate matter (PM). Using ground  
19 observations from 40 cities, GEOS-FP meteorology, CEDS emissions, and monthly LightGBM  
20 models, this study assesses the drivers of PM<sub>2.5</sub> and PM<sub>10</sub> across the Beijing–Tianjin–Hebei (BTH)  
21 and Yangtze River Delta (YRD) regions during 2015–2022. The models demonstrate strong  
22 predictive skill ( $R/R^2 = 0.82/0.67$  for PM<sub>2.5</sub> and  $0.81/0.65$  for PM<sub>10</sub>), with consistently high  
23 performance across cities. Both pollutants exhibit significant decreasing trends over the study period.  
24 Counterfactual experiments show that emission reductions overwhelmingly dominate these  
25 improvements. PM<sub>2.5</sub> emission driven changes intensify from  $-9.1 \mu\text{g m}^{-3}$  in 2016 to  $-31.4 \mu\text{g m}^{-3}$   
26 in 2022, while PM<sub>10</sub> reductions strengthen from  $-9.8$  to  $-42.9 \mu\text{g m}^{-3}$ . Meteorology driven  
27 contributions appear as positive net anomalies at the interannual scale (approximately  $+2-4 \mu\text{g m}^{-3}$   
28 for PM<sub>2.5</sub> and  $+0.5-3 \mu\text{g m}^{-3}$  for PM<sub>10</sub>), indicating that air quality improvements were achieved  
29 despite year to year meteorological influences. SHAP attribution highlights 2-m air temperature

30 (T2M), humidity (QV2M), and key precursors (SO<sub>2</sub>\_total, BC\_total, NO<sub>x</sub>\_total as dominant  
31 predictors. Interaction diagnostics further indicate that meteorological conditions modulate the  
32 effectiveness of precursor emissions, without implying direct causal mechanisms. These results  
33 provide a comprehensive data driven assessment of the factors shaping PM evolution in two major  
34 urban clusters of China.

## 35 **1 Introduction**

36 Particulate matter (PM) is a significant air pollutant and is also a critical research topic in  
37 environmental science due to its diverse sources, complex chemical composition, and profound  
38 impacts on human health (Zhang et al., 2022a). Classified by aerodynamic diameter, PM<sub>2.5</sub> (fine  
39 particles,  $\leq 2.5\mu\text{m}$ ) and PM<sub>10</sub> (inhalable particles,  $\leq 10\mu\text{m}$ ) exert differential impacts on ecosystems  
40 and human health owing to their distinct physicochemical properties and environmental behaviors  
41 (WHO, 2021). Fine particles (PM<sub>2.5</sub>) penetrate deep into the lungs and cross the alveolar–blood  
42 barrier into systemic circulation, while coarser particles (PM<sub>10</sub>) deposit predominantly in the upper  
43 respiratory tract (Fu et al., 2024). Chronic exposure to PM<sub>2.5</sub> is linked to respiratory/cardiovascular  
44 diseases, declines in lung function, and impairment of the immune system (Franklin et al., 2008;  
45 Kioumourtzoglou et al., 2016), Whereas PM<sub>10</sub> aggravates asthma , chronic obstructive pulmonary  
46 disease (COPD), and other respiratory conditions (Seaton et al., 1995). As two pivotal economic  
47 engines of China, the Beijing-Tianjin-Hebei (BTH) and Yangtze River Delta (YRD) regions,  
48 characterized by dense industrial clusters and populations, generate substantial industrial and  
49 transportation emissions, with high-intensity production and daily activities resulting in long-  
50 standing composite air pollution dominated by PM<sub>2.5</sub>, PM<sub>10</sub>, and ozone (Dai et al., 2021, 2023),  
51 posing persistent threats to human health and urban livability. Furthermore, PM pollution acidifies  
52 aquatic environments, disrupts ecosystem balance, degrades soils, and contributes to acid rain and  
53 terrestrial biosphere damage (Dominici et al., 2014; Jerrett, 2015).

54 To address severe air pollution problem, Chinese government implemented the Air Pollution  
55 Prevention and Control Action Plan (State Council of the People’s Republic of China, 2013) and the  
56 Three-Year Action Plan for Winning the Blue Sky Defense Battle (State Council of the People’s  
57 Republic of China, 2018).These initiatives led to substantial reductions in PM concentrations  
58 nationwide (Song et al., 2023). However, China's current Ambient Air Quality Standards (GB 3095-

59 2012) stipulate Grade II annual mean limits of  $35 \mu\text{g m}^{-3}$  for  $\text{PM}_{2.5}$  and  $70 \mu\text{g m}^{-3}$  for  $\text{PM}_{10}$ , which  
60 significantly exceed the updated WHO guidelines (AQG 2021).

61 The dynamics of PM are shaped by anthropogenic precursor emissions—sulfur dioxide ( $\text{SO}_2$ ),  
62 nitrogen oxides ( $\text{NO}_x$ ), and ammonia ( $\text{NH}_3$ )—together with meteorological factors such as  
63 temperature, humidity, precipitation, pressure, and wind (Xiao et al., 2021). In addition to these  
64 inorganic precursors, volatile organic compounds (VOCs) also play an important role in secondary  
65 aerosol formation, particularly through pathways leading to secondary organic aerosols, as  
66 recognized in numerous atmospheric chemistry studies.  $\text{PM}_{2.5}$  originates predominantly from traffic  
67 and industrial emissions, combustion processes (e.g., cooking, biomass burning), and secondary  
68 formation via atmospheric oxidation to sulfate, nitrate, and organic aerosols (Zhang et al., 2015).  
69  $\text{PM}_{10}$  also includes coarse particles from fugitive dust (construction, agriculture) and secondary  
70 coarse-mode particulates (Wu and Huang, 2021). The  $\text{SO}_2$ ,  $\text{NO}_x$ , and  $\text{NH}_3$  in the atmosphere can be  
71 converted into secondary inorganic aerosols, which significantly regulate PM concentrations (Ding  
72 et al., 2019; Feng et al., 2021). Meteorological parameters, such as temperature, relative humidity,  
73 precipitation, pressure, and wind, critically influence PM generation, dispersion, and removal  
74 (Leung et al., 2018; Zhao et al., 2013). For instance, elevated temperatures accelerate  $\text{SO}_x/\text{NO}_x$   
75 oxidation rates and fine PM formation (Chen et al., 2022). High humidity promotes particle  
76 hygroscopic growth, gas-to-particle conversion (e.g., secondary organic aerosols), and wet  
77 deposition, thereby altering PM size distribution and lifetime. These PM-meteorology interactions  
78 exhibit region- and year-specific nonlinear characteristics (Shen et al., 2017), challenging  
79 conventional linear modeling approaches (Zhao et al., 2018)

80 Machine learning (ML), with its capacity to capture complex, nonlinear relationships, has  
81 emerged as a powerful tool for atmospheric pollution research (Yin et al., 2022b). ML enhances  
82 source apportionment accuracy through multi-source data integration (meteorological, emission,  
83 socioeconomic), high-dimensional pattern recognition, and real-time adaptive analysis, enabling  
84 identification of complex pollutant interactions (Peng et al., 2024). For  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$  studies, ML  
85 facilitates quantitative disentanglement of meteorological and emission contributions, elucidates  
86 source-receptor relationships, and informs targeted mitigation strategies.

87 This study employs the LightGBM framework to quantify the drivers of  $\text{PM}_{2.5}$  and  $\text{PM}_{10}$

88 variability across the BTH and YRD regions during 2015–2022. By leveraging LightGBM’s  
89 capability to model nonlinear emission–meteorology–pollution interactions and its efficiency on  
90 multi-year, multi-city datasets, the analysis aims to identify the dominant factors governing regional  
91 air-quality evolution. The structure of this paper is as follows. Section 2 introduces the datasets used  
92 in this study, including national ground-based PM observations (2.1), GEOS-FP meteorological  
93 reanalysis fields (2.2), and the CEDS anthropogenic emission inventory (2.3). Section 3 describes  
94 the methodological framework. Section 3.1 presents the extraction of city-level meteorological and  
95 emission variables from gridded datasets. Section 3.2 outlines the LightGBM modeling workflow,  
96 including the feature-engineering strategy (3.2.1), the model training and leave-one-year-out cross-  
97 validation procedure (3.2.2), and the SHAP-based interpretation approach (3.2.3). The interannual  
98 trend estimation method is detailed in Section 3.3, and the counterfactual framework used to  
99 separate meteorological and emission contributions is introduced in Section 3.4. Section 4 reports  
100 the main results, including the interannual evolution of PM<sub>2.5</sub> and PM<sub>10</sub> (4.1), machine-learning  
101 model performance and key predictor importance (4.2), and the quantified meteorological and  
102 emission contributions (4.3). Section 5 provides a broader discussion of the identified driving  
103 mechanisms in the context of chemical formation pathways and emission-control policies. Finally,  
104 Section 6 summarizes the key findings and presents implications for future air-quality management  
105 and research.

## 106 **2 Data**

### 107 **2.1 Observational data from national monitoring sites**

108 The ground-level air pollutant data for the YRD and BTH regions were acquired from the  
109 China National Environmental Monitoring Center (CNEMC) network (<https://www.cnemc.cn/>, last  
110 accessed: December 31, 2022), comprising hourly measurements of PM<sub>2.5</sub>, PM<sub>10</sub>, SO<sub>2</sub>, NO<sub>2</sub>, CO,  
111 and O<sub>3</sub> concentrations from 2015 to 2022. Observations from multiple monitoring stations within  
112 the same city were averaged to derive city-level pollutant concentrations (site-specific details are  
113 provided in Table S1 and Fig. S1). The monitoring network includes 80 stations in the BTH region,  
114 covering major cities and areas in Beijing, Tianjin, and Hebei Province, and 197 stations in the YRD  
115 region, spanning Shanghai, Jiangsu, Zhejiang, and adjacent provinces. To avoid ambiguity between  
116 two cities sharing the same English spelling “Taizhou”, Taizhou City in Jiangsu Province is denoted

117 as TaizhouJS, while Taizhou City in Zhejiang Province is denoted as TaizhouZJ throughout this  
118 study.

119 All national monitoring stations strictly follow the Technical Specifications for Automatic  
120 Ambient Air Quality Monitoring (Ministry of Environmental Protection of China, 2013a), ensuring  
121 standardized field operation and quality control procedures. City-level PM<sub>2.5</sub> and PM<sub>10</sub>  
122 concentrations are released in accordance with the national reference gravimetric method (Ministry  
123 of Environmental Protection of China, 2011), which provides the calibration and traceability  
124 framework for automated particulate measurements across the monitoring network. Gaseous  
125 pollutants (SO<sub>2</sub>, NO<sub>2</sub>, CO, and O<sub>3</sub>) are measured using ultraviolet fluorescence, chemiluminescence,  
126 non-dispersive infrared absorption, and ultraviolet photometric analysis, respectively, following the  
127 national specifications for continuous gaseous monitoring (Ministry of Environmental Protection of  
128 China, 2013b). These unified procedures ensure accuracy, comparability, and long-term stability of  
129 pollutant observations.

## 130 **2.2 GEOS-FP meteorological data**

131 Meteorological fields for 2015–2022 were obtained from the GEOS Forward Processing  
132 (GEOS-FP) product (<http://geoschemdata.wustl.edu/ExtData/>, last accessed: 31 December 2020) at  
133 a spatial resolution of 0.25° × 0.3125°. GEOS-FP provides hourly assimilated meteorological  
134 variables with relatively high spatial and temporal resolution, enabling refined characterization of  
135 mesoscale atmospheric processes over the BTH and YRD regions. Its near–real-time data  
136 assimilation framework has been widely applied in regional atmospheric pollution and transport  
137 studies, supporting accurate representation of dynamic meteorological conditions (Yin et al., 2021b,  
138 2022a, b). Its near–real-time data assimilation system further improves the accuracy of reanalysis-  
139 based meteorological fields and enhances representation of dynamic atmospheric processes (Sun et  
140 al., 2021a, b; Yin et al., 2019, 2020, 2021a). The meteorological parameters used in this study  
141 include total cloud fraction (CLDTOT), precipitation flux (PRECTOT), 2-m specific humidity  
142 (QV2M), 2-m air temperature (T2M), sea-level pressure (SLP), surface downward shortwave  
143 radiation (SWGDN), and 10-m zonal (U10M) and meridional (V10M) wind components.

## 144 **2.3 CEDS emission inventory**

145 Anthropogenic emission data for 2015–2022 were obtained from the Community Emissions

146 Data System (CEDs), which provides monthly mean fluxes at a spatial resolution of  $0.5^\circ \times 0.5^\circ$ . In  
 147 this study, we use emissions of  $\text{CH}_2\text{O}$ ,  $\text{CO}$ ,  $\text{NH}_3$ ,  $\text{NO}_x$ ,  $\text{SO}_2$ ,  $\text{BC}$ ,  $\text{OC}$ , and paraffinic reactive primary  
 148 emissions (PRPE). Emissions were further categorized into eight sectors: non-combustion  
 149 agriculture, energy transformation and extraction, industrial combustion and processes, surface  
 150 transportation, residential and commercial fuel use, solvents, waste treatment and disposal, and  
 151 international shipping. Anthropogenic emission data for 2015–2022 were derived from the CEDs,  
 152 a global inventory providing temporally resolved sector-specific emissions. The CEDs framework  
 153 supports climate change projections and quantifies human-driven interactions between air pollutants  
 154 and climate systems, critical for assessing health and ecosystem impacts.

### 155 **3 Methodology**

#### 156 **3.1 Emission and meteorological data extraction**

157 City-level emission and meteorological variables were derived from gridded emission  
 158 inventories and meteorological reanalysis datasets. For each city, polygon boundaries were obtained  
 159 from the GADM Level-2 shapefile, and all grid cells whose centers fell within the city polygon were  
 160 identified.

161 For emission data, which are expressed as surface fluxes ( $\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ ), the city-scale total  
 162 emission  $E_{\text{city}}(t)$  was obtained by physical integration over all intersecting grid cells:

$$163 \quad E_{\text{city}}(t) = \sum_{i,j} F_{ij}(t) A_{ij} r_{ij} \quad (1)$$

164 where  $F_{ij}(t)$  is the emission flux of grid cell  $(i, j)$  at time  $t$ ;  $A_{ij}$  is the nominal grid-cell  
 165 area; and  $r_{ij}$  is the fractional overlap between the grid cell and the city polygon.

166 This area-weighted integration converts surface fluxes into physically consistent city-total  
 167 emissions.

168 For meteorological variables, which represent intensive state quantities such as near-surface  
 169 temperature (T2M), specific humidity (QV2M), and wind components (U10M/V10M), city-level  
 170 averages were computed as the arithmetic mean of all grid-cell centers located inside the city  
 171 polygon:

$$172 \quad X_{\text{city}}(t) = \frac{1}{N} \sum_{i,j} X_{ij}(t) \quad (2)$$

173 where  $X_{ij}(t)$  is the variable value in cell  $(i, j)$ , and  $N$  is the number of valid grid cells within  
 174 the city.

175 This center-based averaging is computationally efficient and provides a representative estimate  
 176 of the mean meteorological condition. Because the study region lies mainly in the mid-to-low  
 177 latitudes of China, where grid-cell area variation with latitude is minor, this approximation  
 178 introduces negligible bias compared with full area-weighted averaging.

179 Both extraction procedures ensure spatial consistency between emission and meteorological  
 180 datasets and yield temporally continuous, city-level time series for subsequent modeling.

## 181 **3.2 LightGBM modeling**

182 Light Gradient Boosting Machine (LightGBM) is an efficient and scalable implementation of  
183 gradient boosting, extensively applied to regression, classification, and ranking tasks (Yin et al.,  
184 2021c). By using a histogram-based decision-tree algorithm, the LightGBM model drastically  
185 reduces both computation time and memory usage compared to traditional gradient-boosting  
186 methods such as XGBoost (Bian et al., 2023; Zhang et al., 2017). It supports the direct handling of  
187 categorical variables without one-hot encoding, improving efficiency when processing high-  
188 dimensional datasets. During training, LightGBM grows trees in a leaf-wise (best-first) manner,  
189 generating deeper splits along the branch that achieves the largest loss reduction.

190 In contrast, XGBoost and classical Gradient Boosting Decision Trees (GBDT) use a level-wise  
191 growth strategy, which provides stability but can become computationally slower for large-scale  
192 data. LightGBM also offers extensive hyperparameter controls, such as maximum tree depth,  
193 minimum data in leaf, and feature fraction, to balance model complexity and generalization (Ke et  
194 al., 2017). Due to its high predictive accuracy, efficient splitting mechanism, and robust  
195 computational capability, LightGBM has become one of the most widely used gradient-boosting  
196 frameworks in environmental modeling (Liu et al., 2023; Wang et al., 2022; Zhang et al., 2022b).

197 Prior to model training, all hourly emission and meteorological inputs were aggregated to  
198 monthly means, and the LightGBM models were trained on these monthly-resolved datasets. This  
199 temporal aggregation ensured that all variables shared the same temporal frequency, preventing  
200 inconsistencies between hourly and monthly features. It also matched the model input scale with  
201 the monthly trend analysis period (2015–2022), thereby avoiding time-scale mismatches between  
202 predictor variables and the evaluation framework.

203 The model performance was evaluated using three widely recognized regression metrics: the  
204 correlation coefficient ( $R$ ), indicating the linear relationship between predicted and observed  
205 concentrations. the coefficient of determination ( $R^2$ ), measuring the proportion of variance in  
206 observations explained by the model; and the root-mean-square error (RMSE), representing the  
207 average magnitude of prediction errors. Higher  $R$  and  $R^2$  and lower RMSE indicate stronger  
208 predictive ability.

### 209 **3.2.1 Feature Engineering**

210 To ensure interpretability and reduce redundancy, all anthropogenic emissions were aggregated  
 211 into a single total value for each species (e.g., NOx\_total, SO2\_total). Rather than treating sector-  
 212 resolved emissions separately, emissions from all sectors were summed into one unified variable  
 213 per pollutant. This consolidation reduces feature dimensionality, mitigates multicollinearity among  
 214 sectoral components, and preserves the dominant emission signal, resulting in a more compact and  
 215 interpretable model structure.

216 To incorporate temporal information, three seasonal descriptors were introduced: (1) a pair of  
 217 harmonic terms (month\_sin and month\_cos) representing the cyclic annual progression; (2) a  
 218 categorical season indicator to reflect broad seasonal regimes. These features provide smooth,  
 219 physically meaningful representations of seasonality without imposing sharp discontinuities  
 220 between months.

221 In addition, two derived emission indicators were constructed to characterize temporal  
 222 variability and multi-scale coupling:

223 Seasonal difference (sdiff) represents interannual seasonal changes over a 12-month interval.  
 224 For each feature  $x$ , the seasonal difference is defined as:

$$225 \quad \text{sdiff}_t(x) = x_t - x_{t-12} \quad (3)$$

226 where  $x_t$  denotes the monthly mean value at time  $t$ . This feature highlights annual-scale variations  
 227 and helps the model capture year-to-year emission variability under identical seasonal conditions.

228 Rolling detrended residual (detr) represents short-term deviations from a 12-month moving-  
 229 mean trend. It is expressed as:

$$230 \quad \text{detr}_t(x) = x_t - \mu_t^{(12)}(x) \quad (4)$$

$$231 \quad \mu_t^{(12)}(x) = \frac{1}{k_t} \sum_{i=t-11}^t x_i \cdot 1_{\{x_i \text{ valid}\}}, \quad k_t \geq 3 \quad (5)$$

232 Where  $\mu_t^{(12)}(x)$  is the rolling mean computed over the past 12 months, requiring at least three  
 233 valid data points ( $k_t \geq 3$ ). This feature isolates short-term fluctuations by removing low-frequency  
 234 seasonal trends.

235 In practice, the final predictor set comprised 35 variables: eight monthly meteorological  
 236 parameters, eight species-level aggregated emission totals (e.g., NOx\_total, SO2\_total, BC\_total),  
 237 three temporal descriptors (month\_sin, month\_cos, season), and two derived indicators (sdiff and

238 detr) for each emission species. Pollutant concentrations and explicit calendar identifiers (year,  
239 month, date) were excluded from the input space to avoid information leakage and to ensure  
240 consistent temporal treatment. A complete list of variables used in the model is provided in Table  
241 S2.

### 242 **3.2.2 Model Training and Cross-Validation Procedure**

243 Before model fitting, all months with missing PM<sub>2.5</sub> or PM<sub>10</sub> observations were removed to  
244 ensure consistency between predictors and targets. The derived emission features (sdiff and detr)  
245 inherently contain missing values during their initial 12-month computation window; these were  
246 addressed using a simple mean-imputation strategy applied within each training fold. Specifically,  
247 missing entries for each feature were replaced with the corresponding feature mean computed solely  
248 from the training subset of that fold, thereby preventing temporal leakage and ensuring that  
249 imputation relied exclusively on information available before prediction.

250 The LightGBM model was independently trained for each city, pollutant type (PM<sub>2.5</sub> and PM<sub>10</sub>),  
251 and cross-validation fold. A leave-one-year-out (LOGO) cross-validation scheme was adopted,  
252 whereby data from a single year were held out for testing while the remaining seven years were  
253 used for training. This process was repeated sequentially so that each year between 2015 and 2022  
254 served once as the held-out test period. The resulting predictions thus constitute out-of-sample  
255 estimates for every year, providing a conservative and temporally robust assessment of model  
256 generalizability and avoiding within-year information leakage.

257 Within each outer leave-one-year-out (LOGO) evaluation split, hyperparameters were tuned  
258 using only the training years, with the held-out test year completely excluded from all tuning steps.  
259 Specifically, a randomized search was conducted on the training subset to sample 10 candidate  
260 configurations from predefined distributions, and candidates were ranked by their cross-validated  
261 negative mean squared error (MSE) computed solely within the training data. The selected  
262 hyperparameters (learning\_rate, n\_estimators, num\_leaves, max\_depth, min\_child\_samples,  
263 subsample, colsample\_bytree, reg\_alpha, and reg\_lambda) were then fixed and used to refit the  
264 model on the full training subset of that outer split before generating predictions for the held-out  
265 year. This nested evaluation design ensures that both model fitting and hyperparameter selection  
266 rely exclusively on information available prior to prediction, thereby preventing temporal leakage.

267 A consolidated summary of the optimized hyperparameter ranges is presented in Table S3.

### 268 3.2.3 SHAP Interpretation Analysis

269 To interpret the LightGBM outputs and quantify the contribution of individual predictors, the  
270 Shapley Additive explanations (SHAP) framework was applied. For each trained model, SHAP  
271 values  $s_{i,j}$  represent the marginal contribution of feature  $j$  to the prediction of sample  $i$ . They are  
272 derived from cooperative game theory and satisfy the local additivity principle:

$$273 \quad f(x_i) = E[f(x)] + \sum_{j=1}^m s_{i,j} \quad (6)$$

274 where  $f(x_i)$  is the model prediction for sample  $i$ , and  $E[f(x)]$  is the expected model  
275 output over all samples.

276 Because this study involves multiple cities across the BTH and YRD regions, a unified regional  
277 SHAP ranking was derived by weighting each SHAP importance of city by its corresponding  
278 number of valid monthly samples. The weighted importance for feature  $j$  is defined as:

$$279 \quad I_j^{(w)} = \frac{\sum_{c=1}^N I_{j,c} n_c}{\sum_{c=1}^N n_c} \quad (7)$$

280 Here  $I_{j,c}$  is the SHAP-based importance of feature  $j$  in city  $c$ , and  $n_c$  is the number of  
281 valid samples for that city,  $N$  is the total number of cities.

282 This weighting strategy ensures that cities with longer and more complete observational  
283 records have proportionally greater influence on the regional-level interpretability.

### 284 3.3 Interannual trend analysis method

285 To quantify the interannual trends of PM<sub>2.5</sub> and PM<sub>10</sub> concentrations from 2015 to 2022, a linear  
286 regression model was employed in this study. For each city, the relationship between annual mean  
287 concentration  $y$  and year  $x$  was modeled as:

$$288 \quad y = \beta_0 + \beta_1 x + \epsilon \quad (8)$$

289 where  $\beta_0$  represents the intercept (baseline concentration), and  $\epsilon$  denotes the error term. The  
290 slope  $\beta_1$ , reflecting the annual rate of concentration change, was estimated via the ordinary least  
291 squares (OLS) method. Specifically, the parameters were optimized by minimizing the residual sum  
292 of squares (RSS):

$$293 \quad \arg \min_{\beta_0, \beta_1} \sum_{i=1}^n (y_i - (\beta_0 + \beta_1 x_i))^2 \quad (9)$$

294 where  $n$  is the sample size (e.g.,  $n=6$  for the period 2015–2022),  $x_i$  denotes the year, and  $y_i$   
295 represents the corresponding annual mean concentration.

296 The slope  $\beta_1$  was derived as:

$$297 \quad \beta_1 = \frac{\text{Cov}(x, y)}{\text{Var}(x)} \quad (10)$$

298 The sign of  $\beta_1$  indicates the direction of concentration trends (negative for decreasing,  
299 positive for increasing), while its absolute value quantifies the magnitude of change.

### 300 **3.4 Methodology for disentangling meteorological and emission contributions**

301 To quantify meteorological and anthropogenic emission contributions, the trained models were  
302 applied in a parallel prediction experiment. For each year from 2016 to 2022, anthropogenic  
303 emission features were held fixed at their 2015 levels, while meteorological and temporal predictors  
304 were retained at their actual contemporaneous values. This produced a counterfactual concentration  
305 series driven solely by meteorological variability. This yielded pollutant concentrations driven  
306 solely by meteorological variations (denoted as  $ML_{2022\text{met}}$  for 2022). The contribution metrics  
307 related to 2015 were calculated as follows:

308 Meteorological contribution ( $ML_{2022\text{met}}$ ):

$$309 \quad ML_{2022\text{met}} = ML_{15-22} - ML_{2015} \quad (11)$$

310  $ML_{15-22}$  is the non-emission condition unchanged, the emission condition is fixed as the model  
311 prediction result in 2015, and  $ML_{2015}$  is the model prediction result with unchanged meteorological  
312 and emission conditions.

313 Emission contribution ( $ML_{2022\text{emis}}$ ):

$$314 \quad ML_{2022\text{emis}} = (Obs_{2022} - Obs_{2015}) - ML_{2022\text{met}} \quad (12)$$

315  $Obs_{2022}$  and  $Obs_{2015}$ : Observed concentrations in 2022 and 2015, respectively.

## 316 **4 Results**

### 317 **4.1 Interannual trends of ground-level PM<sub>2.5</sub> and PM<sub>10</sub>**

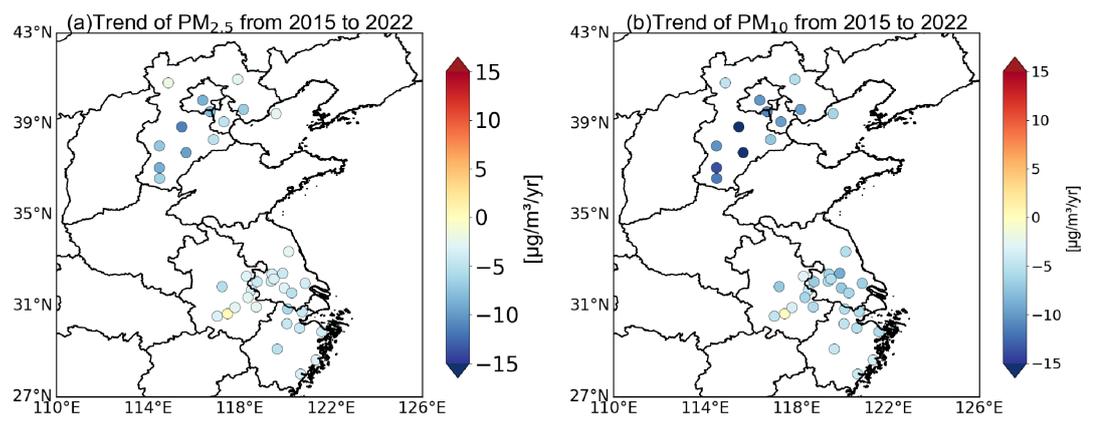
318 Fig. 1 illustrates the interannual evolution of ground-level PM<sub>2.5</sub> and PM<sub>10</sub> concentrations  
319 across the BTH and YRD regions from 2015 to 2022, with corresponding annual mean spatial  
320 distributions shown in Fig. S2–S3. Both pollutants exhibit persistent downward trajectories across  
321 nearly all cities. The monthly time series and associated OLS trend lines (Fig. S4) further confirm  
322 that these declines remain stable throughout the study period, with the most pronounced reductions

323 occurring before 2020. Statistical diagnostics from linear regression (Table S4) indicate that most  
324 cities display significant negative trends ( $p < 0.05$ ), underscoring the robustness and consistency of  
325 these decreases.

326 For  $PM_{2.5}$ , all cities show negative trends, with annual rates ranging from approximately  $-1.53$   
327 to  $-9.46 \mu\text{g m}^{-3} \text{ yr}^{-1}$ . Within BTH, the most rapid declines occur in Baoding ( $-9.46 \pm 0.74 \mu\text{g m}^{-3}$   
328  $\text{yr}^{-1}$ ), Hengshui ( $-8.36 \pm 0.67 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ), and Xingtai ( $-8.02 \pm 0.56 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ), while Chengde  
329 ( $-1.98 \pm 0.35 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ) and Zhangjiakou ( $-2.00 \pm 0.42 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ) exhibit more modest declines.  
330 In the YRD region, substantial decreases are observed in Huzhou ( $-5.69 \pm 1.22 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ), Hefei  
331 ( $-4.83 \pm 0.33 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ), and Chuzhou ( $-4.28 \pm 1.12 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ), whereas Zhoushan ( $-2.11 \pm$   
332  $0.14 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ) and Taizhou-ZJ ( $-2.72 \pm 0.26 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ) experience comparatively smaller  
333 reductions. These regional contrasts closely align with the initial concentration levels shown in Fig.  
334 S2, where inland BTH cities began with substantially higher baselines (e.g., Baoding and Hengshui  
335 exceeded  $105$  and  $98 \mu\text{g m}^{-3}$  in 2015), enabling larger absolute declines.

336 For  $PM_{10}$ , all cities also exhibit significant decreases, with annual trends ranging from roughly  
337  $-1.84$  to  $-13.79 \mu\text{g m}^{-3} \text{ yr}^{-1}$ . The largest reductions occur in BTH, particularly Hengshui ( $-13.79 \pm$   
338  $1.82 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ), Baoding ( $-13.07 \pm 1.46 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ), and Xingtai ( $-12.76 \pm 1.29 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ). In  
339 contrast, Chizhou ( $-1.84 \pm 2.03 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ) and Zhoushan ( $-2.77 \pm 0.30 \mu\text{g m}^{-3} \text{ yr}^{-1}$ ) show the  
340 smallest declines, and the  $PM_{10}$  trend in Chizhou is not statistically significant ( $p = 0.40$ ), consistent  
341 with its minimal decline rate. As with  $PM_{2.5}$ , cities starting with the highest  $PM_{10}$  levels—such as  
342 Baoding ( $174.6 \mu\text{g m}^{-3}$  in 2015) and Hengshui ( $175.9 \mu\text{g m}^{-3}$ )—exhibit the steepest reductions,  
343 whereas cities with lower initial levels (e.g., Zhoushan at  $\sim 46.8 \mu\text{g m}^{-3}$ ) show correspondingly  
344 smaller absolute decreases.

345 Overall, both  $PM_{2.5}$  and  $PM_{10}$  decrease more rapidly in BTH than in YRD, reflecting regional  
346 differences in initial emissions, industrial structure, and the intensity of mitigation policies. The  
347 widespread statistical significance of the trends (Table S4) supports the conclusion that both regions  
348 experienced sustained and robust improvements in air quality during 2015–2022.



349

350 **Fig. 1** Interannual variation trends of PM<sub>2.5</sub> (a) and PM<sub>10</sub> (b) in each city during 2015-2022.

### 351 4.2 Machine learning model performance and variable importance

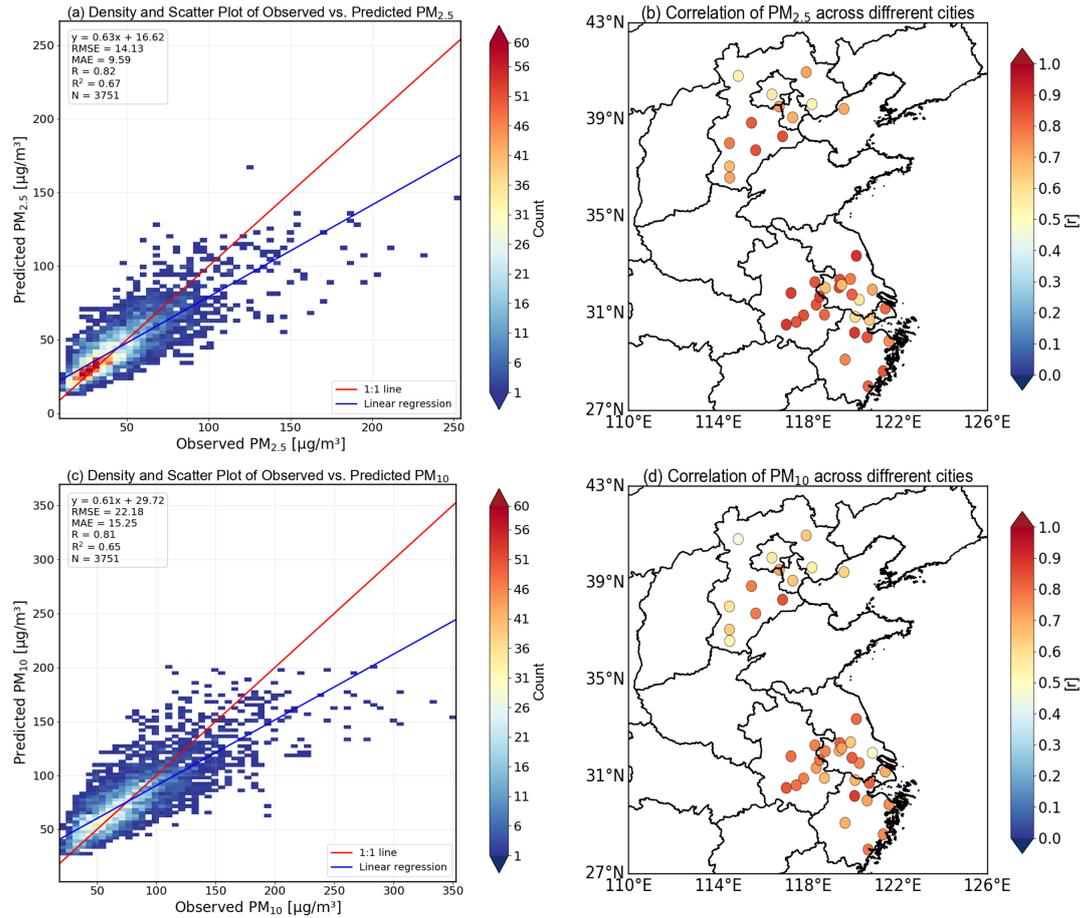
352 Fig. 2 summarizes the predictive performance of the LightGBM models for PM<sub>2.5</sub> and PM<sub>10</sub>  
 353 across all cities. The density scatterplots (Fig. 2a and 2c) show good agreement between predictions  
 354 and observations, with overall R/R<sup>2</sup> values of 0.82/0.67 for PM<sub>2.5</sub> and 0.81/0.65 for PM<sub>10</sub>,  
 355 respectively. City-level performance (Fig. 2b and 2d) further indicates that the majority of cities  
 356 achieve correlation coefficients exceeding 0.70, as detailed in Table S5. While the predictive skill  
 357 varies among cities, particularly for PM<sub>10</sub>, such variability is expected given the stronger influence  
 358 of coarse particle processes and regional heterogeneity. Importantly, subsequent robustness analyses  
 359 (Fig.S5, Fig.S6) confirm that this variability does not arise from model overfitting or instability, but  
 360 rather reflects intrinsic differences in local PM dynamics across urban environments.

361 Fig. 3 summarizes the SHAP-based overall feature importance. For PM<sub>2.5</sub>, although T2M ranks  
 362 as the single most influential variable in Fig. 3a, the broader pattern shows that emission-related  
 363 predictors dominate the upper portion of the importance distribution, with SO<sub>2</sub>\_total, BC\_total, and  
 364 NO<sub>x</sub>\_total collectively contributing a larger share than any single meteorological factor. This  
 365 implies that PM<sub>2.5</sub> variability during 2015–2022 remains strongly controlled by precursor emissions  
 366 and their long-term reductions. In contrast, for PM<sub>10</sub> (Fig. 3b), meteorological variables, particularly  
 367 QV2M and T2M, occupy the highest ranks. It should be noted that PM<sub>10</sub> inherently includes a  
 368 substantial contribution from fine particles (PM<sub>2.5</sub>). As a result, the similarity between the PM<sub>2.5</sub> and  
 369 PM<sub>10</sub> feature importance rankings partly reflects shared driving processes affecting fine-mode  
 370 particles, rather than a limitation of the modeling framework. Within this integrated metric, the

371 elevated importance of humidity and temperature reflects their role in modulating particle mass  
372 through hygroscopic growth, boundary-layer conditions, and seasonally varying resuspension  
373 processes. Precursor emissions such as NO<sub>x</sub>\_total and SO<sub>2</sub>\_total remain influential, but their relative  
374 importance is reduced compared to PM<sub>2.5</sub>.

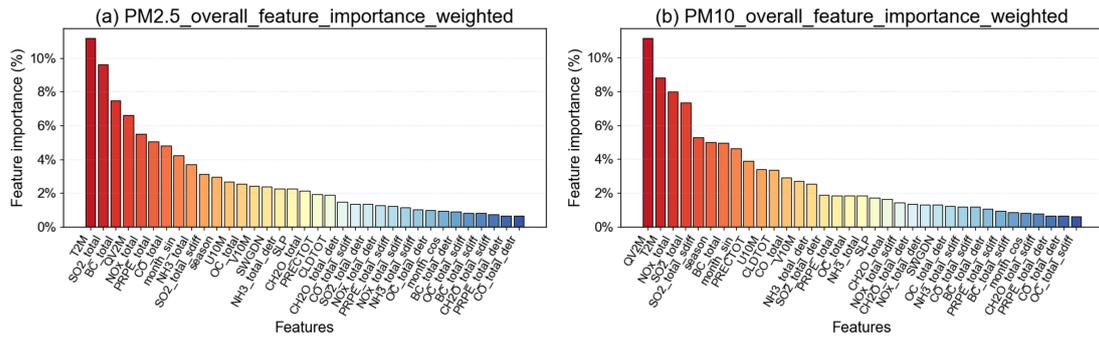
375 To further assess whether the model can distinguish drivers associated with coarse particles,  
376 we additionally analyzed the coarse-mode mass defined as PM<sub>2.5-10</sub>, calculated as PM<sub>10</sub> minus PM<sub>2.5</sub>,  
377 using the same LOGO-based LightGBM modeling and SHAP attribution framework. The resulting  
378 feature importance rankings show systematic differences from those of PM<sub>10</sub>. In particular, the  
379 dominance of humidity- and temperature-related variables and combustion-related emission proxies  
380 is reduced, whereas the relative influence of transport- and removal-related meteorological factors  
381 becomes more pronounced. Detailed results are provided in Supplementary Fig. S7.

382 Region-specific SHAP rankings (Fig. S8) further show that PM<sub>2.5</sub> in the BTH region is more  
383 strongly influenced by SO<sub>2</sub>\_total and BC\_total, whereas the YRD exhibits greater sensitivity to  
384 meteorological conditions such as temperature and humidity. For PM<sub>10</sub>, QV2M remains the  
385 dominant driver in both regions, while precursor importance (e.g., NO<sub>x</sub>\_total) is consistently higher  
386 in BTH. These spatial contrasts reflect differences in emission structures and seasonal behavior  
387 between regions and underscore the need for differentiated control strategies targeting PM<sub>2.5</sub> and  
388 PM<sub>10</sub>.



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**Fig. 2** The density scatter plots of PM<sub>2.5</sub> (a) and PM<sub>10</sub> (c) concentrations observed and predicted, respectively. The correlation of PM<sub>2.5</sub> (b) and PM<sub>10</sub> (d) in each city over BTH and YRD regions, respectively.



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**Fig. 3** The feature importance of rankings of PM<sub>2.5</sub> (a) and PM<sub>10</sub> (b) in ML model prediction, respectively.

### 4.3 Contributions of emissions and meteorology

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The spatial patterns of average emission and meteorological contributions (Fig. 4) reveal clear regional contrasts between northern cities in BTH and southern cities in YRD. For PM<sub>2.5</sub>, emission

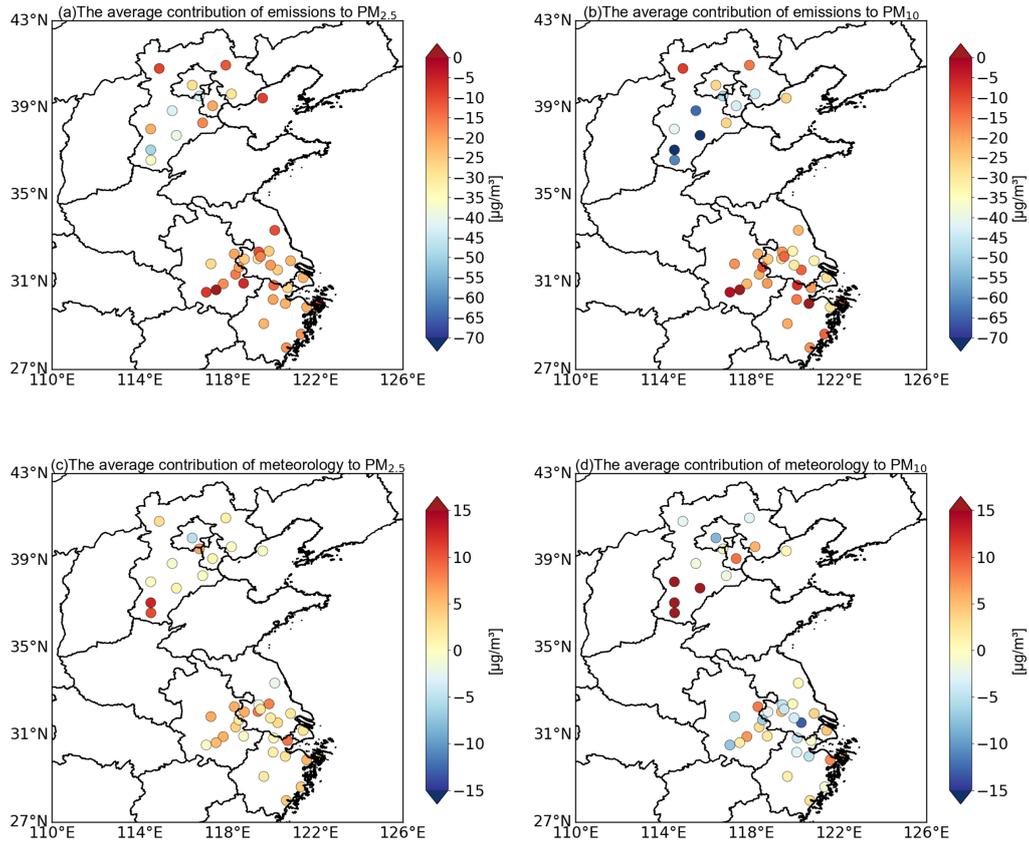
400 contributions are strongly negative across most BTH cities such as Baoding, Hengshui, and Xingtai,  
401 often exceeding  $-30 \mu\text{g m}^{-3}$ , reflecting substantial reductions from historically elevated emission  
402 levels. In contrast, YRD cities such as Suzhou, Hangzhou, and Ningbo display more moderate  
403 negative contributions, typically around  $-10$  to  $-20 \mu\text{g m}^{-3}$ , consistent with their lower baseline  
404 emissions. Meteorological contributions also exhibit north–south gradients: several coastal or  
405 humid subtropical YRD cities (e.g., Ningbo, Wenzhou) show slightly positive meteorological  
406 influences, whereas continental BTH cities (e.g., Beijing, Tianjin) show predominantly negative  
407 effects associated with winter stagnation, shallow boundary layers, and lower humidity. Similar  
408 spatial patterns are observed for  $\text{PM}_{10}$  (Fig. 4b, d), where emission-driven reductions remain larger  
409 in BTH than in YRD, mirroring regional differences in emission intensity.

410 The interannual evolution shown in Fig. 5 further clarifies these relationships. Emission  
411 contributions remain consistently negative from 2016 to 2022 for both pollutants and intensify over  
412 time. For  $\text{PM}_{2.5}$ , the reductions increase from  $-9.13 \mu\text{g m}^{-3}$  in 2016 to  $-31.39 \mu\text{g m}^{-3}$  in 2022,  
413 while  $\text{PM}_{10}$  reductions strengthen from  $-9.77$  to  $-42.91 \mu\text{g m}^{-3}$  over the same period. These  
414 progressively stronger negative contributions are consistent with sustained decreases in major  
415 emission species, including  $\text{NOx}_{\text{total}}$ ,  $\text{SO}_{2\text{total}}$ , and  $\text{BC}_{\text{total}}$ . In contrast, meteorological  
416 contributions remain positive throughout all years, on the order of approximately  $2\text{--}4 \mu\text{g m}^{-3}$  for  
417  $\text{PM}_{2.5}$  and  $0.5\text{--}3 \mu\text{g m}^{-3}$  for  $\text{PM}_{10}$ , indicating that interannual meteorological conditions, on average,  
418 tended to offset part of the emission-driven reductions rather than reinforce them. Although  $\text{PM}_{10}$  is  
419 often considered more responsive to short-term meteorological variability, the smaller meteorology-  
420 driven contributions for  $\text{PM}_{10}$  in Fig. 5 should be interpreted in the context of interannual net effects.  
421 Meteorological influences on  $\text{PM}_{10}$  often involve multiple processes that can act in opposite  
422 directions, such as enhanced resuspension versus enhanced dispersion and removal, leading to  
423 partial cancellation when aggregated at annual timescales. By contrast, meteorological effects on  
424  $\text{PM}_{2.5}$ , particularly those related to temperature and humidity, tend to influence secondary formation  
425 and particle growth in a more directionally consistent manner at the interannual scale. This  
426 consistency allows their impacts to accumulate into a clearer net contribution. As a result, the  
427 relative magnitude of meteorology-driven changes appears larger for  $\text{PM}_{2.5}$  than for  $\text{PM}_{10}$  in Fig. 5,  
428 without implying a weaker overall meteorological sensitivity of  $\text{PM}_{10}$ .

429 Temporal relationships between individual emission indicators and their contributions are  
430 shown in Fig. 6. For PM<sub>2.5</sub>, month-to-month changes in NO<sub>x</sub>\_total, SO<sub>2</sub>\_total, and BC\_total are  
431 strongly correlated with their corresponding emission-driven contributions ( $R = 0.89\text{--}0.95$ ). A  
432 similar correspondence is observed for PM<sub>10</sub> ( $R = 0.89\text{--}0.93$ ). These strong associations indicate  
433 that the estimated emission contributions respond consistently to changes in emission levels, with  
434 higher emissions generally linked to larger positive contributions and lower emissions associated  
435 with weaker contributions. This relationship persists despite the nonlinear structure of the model,  
436 which adjusts contribution magnitudes according to the prevailing meteorological conditions.

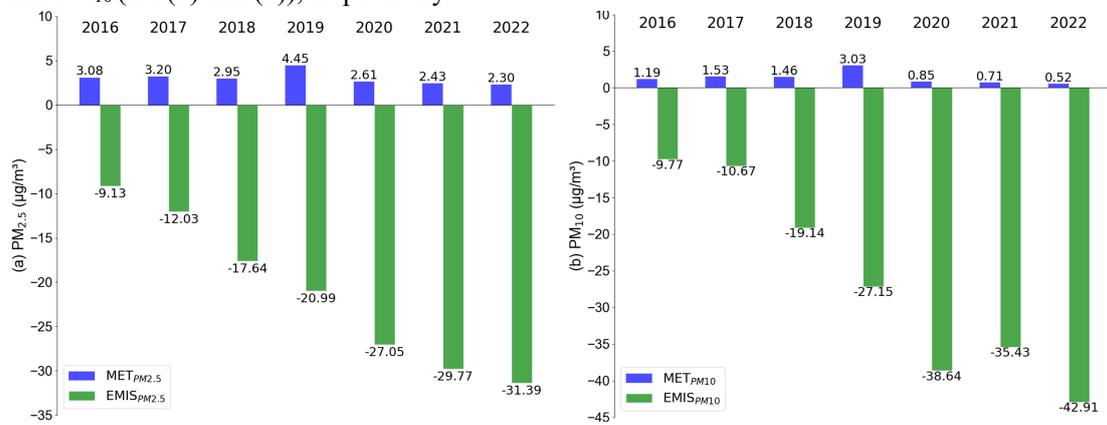
437 In contrast, meteorological drivers show a different temporal behavior (Fig. 7). Temperature  
438 and humidity exhibit strong negative correlations with their meteorology-driven contributions for  
439 both pollutants ( $R \approx -0.96$  to  $-0.98$ ). Importantly, these correlations describe relative contributions  
440 at the interannual scale after emission-driven trends have been removed, rather than direct or  
441 instantaneous responses of particulate matter to meteorological forcing. At this aggregated temporal  
442 scale, years with higher temperature or moisture do not necessarily correspond to larger net  
443 meteorological contributions, because multiple meteorological influences with opposing effects can  
444 partially offset each other. These results therefore point to a scale-dependent influence of  
445 meteorology, rather than contradicting process-based studies that emphasize the role of temperature  
446 and humidity in promoting particle formation at shorter timescales.

447 Taken together, the increasingly negative emission-driven contributions shown in Fig. 5,  
448 combined with relatively modest and predominantly positive net meteorology-driven anomalies at  
449 the interannual scale, indicate that the observed improvements in PM<sub>2.5</sub> and PM<sub>10</sub> during 2016–2022  
450 were mainly driven by sustained emission reductions. Meteorological conditions influenced year-  
451 to-year fluctuations but did not reverse the overall downward trends, highlighting the dominant role  
452 of emission control measures in shaping the long-term evolution of both pollutants.



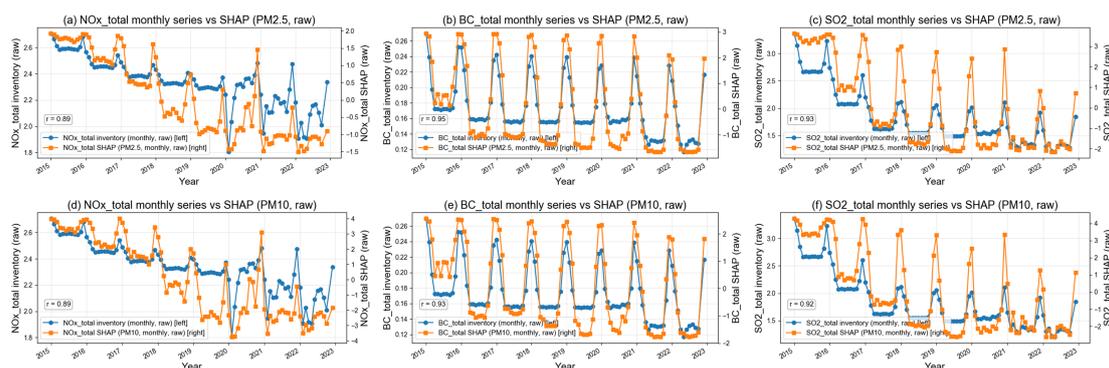
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**Fig. 4** The average contributions of emissions and meteorological variables to  $PM_{2.5}$  (for (a) and (c)) and  $PM_{10}$  (for (b) and (d)), respectively.



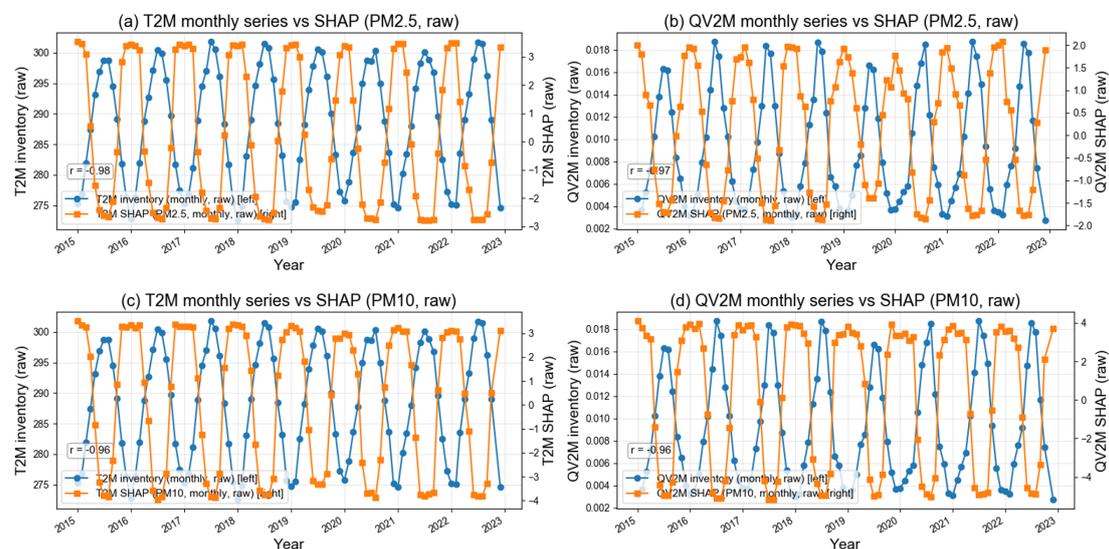
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**Fig. 5** The averaging the emission or meteorological contributions to  $PM_{2.5}$  (a) and  $PM_{10}$  (b) of each year relative to 2015.



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**Fig. 6** Monthly emission inventories of NO<sub>x</sub>\_total, BC\_total, and SO<sub>2</sub>\_total (left y-axis) and their corresponding SHAP values (right y-axis) for PM<sub>2.5</sub> and PM<sub>10</sub> from 2015 to 2022. Panels (a)–(c) show NO<sub>x</sub>\_total, BC\_total, and SO<sub>2</sub>\_total and their SHAP contributions to PM<sub>2.5</sub>, while panels (d)–(f) present the corresponding relationships for PM<sub>10</sub>. Emission inventories and SHAP values are plotted on separate y-axes to account for their different magnitudes.



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**Fig. 7** Monthly meteorological variables (T2M and QV2M; left y-axis) and their corresponding SHAP values (right y-axis) for PM<sub>2.5</sub> and PM<sub>10</sub> from 2015 to 2022. Panels (a)–(b) show the relationships for PM<sub>2.5</sub>, while panels (c)–(d) present the corresponding results for PM<sub>10</sub>. Separate y-axes are used to account for differences in magnitude between meteorological variables and SHAP values.

## 471 5 Discussions

472 To provide additional context for the SHAP-derived attributions, we examine the spatial  
473 distributions and statistical associations of key precursor species. Fig. 8 shows that ambient  
474 concentrations of CO, NO<sub>2</sub>, and SO<sub>2</sub> display pronounced spatial contrasts across the BTH and YRD  
475 regions. Heavy industrial cities such as Tangshan, Xingtai, Handan, and Baoding consistently  
476 exhibit higher levels of these pollutants, whereas several coastal YRD cities (e.g., Zhoushan and  
477 TaizhouZJ) show substantially lower concentrations, reflecting differences in energy structure and

478 industrial activity. These spatial patterns offer a useful background for interpreting the correlation  
479 results summarized in Table 1.

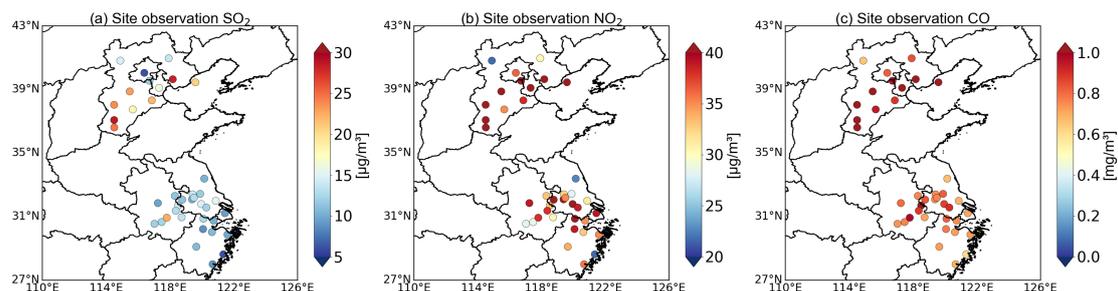
480 Across all cities, CO shows the strongest statistical association with PM<sub>2.5</sub> (mean R ≈ 0.72) and  
481 PM<sub>10</sub> (mean R ≈ 0.62), followed by NO<sub>2</sub> (R ≈ 0.59/0.57) and SO<sub>2</sub> (R ≈ 0.49/0.48). The relatively  
482 strong CO–PM relationships are consistent with their shared combustion-related origins, including  
483 traffic emissions, industrial fuel use, and residential heating, and align with recent multi-platform  
484 and top-down studies reporting tight coupling among CO, NO<sub>x</sub>, carbonaceous aerosols, and  
485 combustion-related PM<sub>2.5</sub>(Tiwari et al., 2025; Wang et al., 2021, 2025). In contrast, the weaker  
486 correlations involving SO<sub>2</sub>, despite elevated concentrations in several northern cities, likely reflect  
487 the long-term effectiveness of desulfurization policies, which have reduced sulfate formation and  
488 altered secondary aerosol composition. NO<sub>2</sub> correlations fall between those of CO and SO<sub>2</sub>,  
489 indicating sustained contributions from traffic and industrial sources while also reflecting evolving  
490 chemical pathways and emission controls. Building on this context, Fig. 9 illustrates the joint  
491 dependence of NO<sub>x</sub>\_total-related SHAP contributions on temperature for PM<sub>2.5</sub>. Lower  
492 temperatures are associated with stronger NO<sub>x</sub>-related contributions, whereas this influence  
493 weakens at higher temperatures. This pattern is consistent with established seasonal behavior in  
494 which cold conditions favor the persistence of nitrate-related particulate matter and reduced  
495 atmospheric mixing, while warmer conditions limit nitrate effectiveness and enhance dilution. Here,  
496 the interaction plot is intended as a diagnostic illustration showing how SHAP-derived contributions  
497 vary across the observed temperature range, rather than as a standalone mechanistic attribution. We  
498 do not present analogous interaction panels for PM<sub>10</sub> or for humidity-related variables. Coarse  
499 particles are more strongly influenced by mechanically driven processes such as dust resuspension  
500 and surface conditions, leading to less systematic chemical responses to temperature. Similarly,  
501 humidity affects multiple competing processes, including hygroscopic growth, aqueous-phase  
502 reactions, boundary-layer suppression, and wet removal, making it difficult to isolate a single,  
503 interpretable interaction at the seasonal scale.

504 Taken together, the analyses in this section are intended to complement the main SHAP-based  
505 attribution results by providing empirical context and internal consistency checks. They suggest that  
506 the statistical relationships identified by the model are broadly compatible with known emission

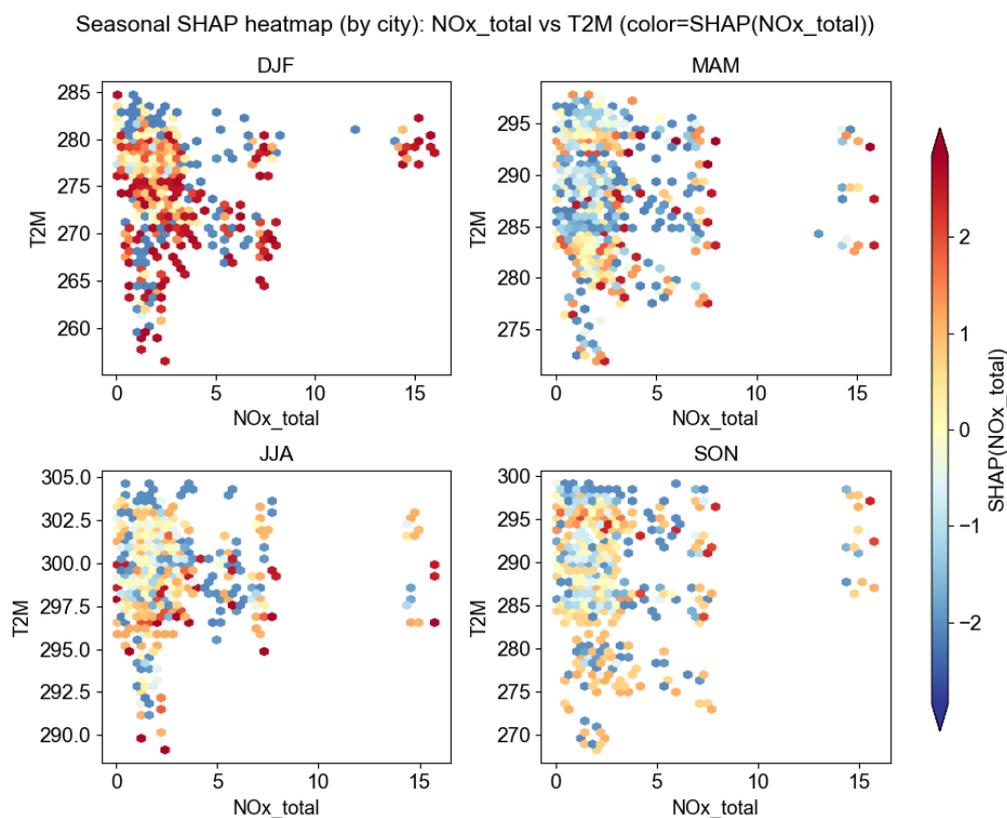
507 structures, chemical regimes, and seasonal behavior, while reinforcing that the primary physical  
 508 interpretation of PM variability is established by the feature importance rankings and contribution  
 509 analyses discussed in earlier sections.

510 **Table 1** The correlation values among SO<sub>2</sub>, NO<sub>2</sub>, CO and PM<sub>2.5</sub> / PM<sub>10</sub>, respectively.

	SO <sub>2</sub>	NO <sub>2</sub>	CO
PM <sub>2.5</sub>	0.49	0.59	0.72
PM <sub>10</sub>	0.48	0.57	0.62



511  
 512 **Fig. 8** The average concentrations of SO<sub>2</sub> (a), NO<sub>2</sub> (b), CO (c), respectively, during 2015 to 2020  
 513 over BTH and YRD regions.



514  
 515 **Fig. 9** Seasonal scatter plots of NO<sub>x</sub>\_total versus T2M across all cities, with point colors representing  
 516 the SHAP contribution of NO<sub>x</sub>\_total. The four panels correspond to the DJF, MAM, JJA, and SON  
 517 seasons.

## 518 **6 Conclusions**

519 This study integrates multi-source emission inventories, GEOS-FP meteorological fields, and  
520 ground-based observations to investigate the drivers of PM<sub>2.5</sub> and PM<sub>10</sub> variability across the BTH  
521 and YRD regions during 2015–2022 using a unified LightGBM modeling and attribution framework.  
522 Both pollutants exhibit clear and statistically significant declines across most cities, supported by  
523 strong model performance under a rigorous leave-one-year-out cross-validation design ( $R/R^2 =$   
524  $0.82/0.67$  for PM<sub>2.5</sub>;  $0.81/0.65$  for PM<sub>10</sub>). The consistent agreement between predictions and  
525 observations across regions and years demonstrates the robustness of the machine-learning-based  
526 representation of emission–meteorology–PM relationships at the interannual scale.

527 A central contribution of this work is the integration of SHAP-based interpretability diagnostics,  
528 which enables transparent attribution while retaining the flexibility of nonlinear learning. The  
529 attribution analyses indicate that anthropogenic emission reductions are the primary driver of the  
530 observed PM improvements. For PM<sub>2.5</sub>, emission-driven decreases intensify from  $-9.13 \mu\text{g m}^{-3}$  in  
531 2016 to  $-31.39 \mu\text{g m}^{-3}$  in 2022, while corresponding reductions for PM<sub>10</sub> strengthen from  $-9.77$  to  
532  $-42.91 \mu\text{g m}^{-3}$ . These progressively larger negative contributions are consistent with sustained  
533 declines in major precursors, including NO<sub>x</sub>, SO<sub>2</sub>, and BC. In contrast, meteorology-driven  
534 contributions appear as comparatively smaller net anomalies at the interannual scale, indicating that  
535 year-to-year meteorological variability modulated PM levels but did not offset the long-term  
536 emission-driven downward trends.

537 The consistency between precursor emissions and their SHAP-derived contributions at the  
538 monthly scale, together with scale-aware temperature and humidity patterns, provides additional  
539 confidence that the attribution results are compatible with established emission structures, seasonal  
540 behavior, and chemical regimes. Rather than serving as direct mechanistic proofs, these diagnostics  
541 offer internal consistency checks that support the interpretability of the machine-learning framework.

542 Several methodological strengths emerge from this study. The use of harmonized monthly  
543 emission totals, multi-scale temporal descriptors (sdiff and detr), and SHAP-based interaction  
544 diagnostics facilitates a physically informed interpretation of complex emission–meteorology  
545 coupling. In addition, the explicit construction of counterfactual predictions provides a transparent  
546 approach for separating meteorological and anthropogenic influences, offering a reproducible

547 pathway for applying machine-learning-based attribution methods to other regions and pollutants.

548       Despite these advantages, some limitations remain. The monthly temporal resolution cannot  
549 fully capture short-term meteorological or chemical processes; uncertainties in bottom-up emission  
550 inventories and meteorological reanalyzes may affect the absolute magnitudes of estimated  
551 contributions; and, as with all data-driven approaches, causal relationships cannot be inferred  
552 directly from statistical associations. Future work will extend this framework to explicitly examine  
553 coarse-mode particles by analyzing PM<sub>2.5-10</sub>, enabling a clearer separation of fine particle and coarse  
554 particle drivers. Additional developments may include higher-frequency observations, expanded  
555 precursor coverage (e.g., VOCs), and hybrid machine learning and chemical transport modeling to  
556 further improve process interpretability and physical fidelity.

557       Overall, this study demonstrates that sustained multi-sector anthropogenic emission reductions,  
558 rather than meteorological variability, primarily explain the observed decreases in PM<sub>2.5</sub> and PM<sub>10</sub>  
559 from 2015 to 2022. The results highlight the value of physically informed machine learning tools,  
560 coupled with SHAP-based interpretability, for diagnosing long term air quality evolution and  
561 supporting emission control strategies grounded in observational evidence.

562

### 563 **Code and data availability**

564 The code and data for this study can be found on <https://doi.org/10.5281/zenodo.17779780>.

565

### 566 **Competing interests**

567 The contact author has declared that none of the authors has any competing interests.

568

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579 2022YFC3700100).

580

#### 581 **Author contributions**

582 HY and YWS designed this study. ZFP wrote the paper with help from HY and YWS. ZFP  
583 contributed to analysis of the data for this study. All co-authors commented on this study.

584

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