

1 **Tracking surface ozone responses to clean air actions under**
2 **a warming climate in China using machine learning**

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19

20 **Abstract.** Surface ozone, a major air pollutant with important implications for human health, ecosystems,
21 and climate, shows long-term trends shaped by both anthropogenic and climatic drivers. Here, we
22 developed a machine learning-based approach, namely the fixed emission approximation (FEA), to
23 disentangle the effects of meteorological variability and anthropogenic emissions on summertime ozone
24 trends in China. We identified three phases of ozone trends corresponding to clean air actions.
25 Anthropogenic emissions drove an approximately $+23.2 \pm 1.1 \mu\text{g m}^{-3}$ increase in summer maximum
26 daily 8-hour average ozone during 2013–2017, followed by an approximately $-4.6 \pm 1.5 \mu\text{g m}^{-3}$ decrease
27 between 2017 and 2020. However, extreme meteorological anomalies, including heatwaves and
28 extended rainfall, emerged as key drivers of ozone variability during 2020–2023. Satellite-derived
29 formaldehyde-to-nitrogen dioxide ratios revealed widespread urban volatile organic compounds-limited
30 regimes, with a shift toward nitrogen oxides-limited sensitivity under the influence of heatwaves. Finally,
31 we assess ozone trends under sustained climate warming from 1970 to 2023 based on the FEA
32 framework. The results indicate a significant climate-driven increase in ozone levels across China's
33 urban agglomerations, underscoring the amplifying role of climate change in ozone pollution.
34 Together, these findings highlight the dual influence of anthropogenic and climatic factors on ozone
35 pollution and emphasize the need for integrated strategies that couple emission mitigation with
36 climate adaptation to effectively manage ozone risks in a warming world.

37 **1 Introduction**

38 Surface ozone (O_3) is a critical air pollutant that poses significant threats to human health (Knowlton et
39 al., 2004), ecosystems (Agathokleous et al., 2020), and climate (Fishman et al., 1979; Hauglustaine et
40 al., 1994). It forms through complex photochemical reactions involving nitrogen oxides (NO_x) and
41 volatile organic compounds (VOCs) in the presence of sunlight (Jacob, 2000; Wang et al., 2017),
42 exhibiting a nonlinear response to its precursors (Guo et al., 2023; Liu and Shi, 2021; Wang et al., 2023a).
43 Controlling ozone pollution remains a global environmental challenge. In recent years, China has
44 implemented a series of national clean air actions, most notably the Air Pollution Prevention and Control
45 Action Plan (2013–2017) and the Three-Year Action Plan for Winning the Blue-Sky War (2018–2020)
46 (Geng et al., 2024; Zhang et al., 2019; Zheng et al., 2018), that have markedly improved air quality,
47 particularly by reducing fine particulate matter ($PM_{2.5}$) (Geng et al., 2024; Zhang et al., 2019). However,
48 surface ozone levels have continued to rise in many regions, raising concerns over the complex drivers
49 of ozone trends and highlighting the need for scientific attribution to guide effective mitigation strategies
50 (Li et al., 2019a; Liu et al., 2023; Wang et al., 2023a; Weng et al., 2022).

51 Long-term ozone variability is jointly influenced by anthropogenic emissions and weather
52 conditions as well as regional climate (Hallquist et al., 2016; Li et al., 2019b; Wang et al., 2022a). While
53 emission controls directly regulate precursor abundance, climate change modulates ozone through
54 chemical feedbacks, meteorological dynamics, and biosphere–atmosphere interactions (Ma and Yin,
55 2021; Xue et al., 2020). Over the past century, global surface temperatures have substantially increased
56 relative to the pre-industrial baseline (1850–1900), driven largely by human activities (IPCC, 2021). In
57 such a warming world, extreme climate anomalies – such as heatwaves and persistent rainfall shifts – are
58 expected to intensify (Li et al., 2025a; Li et al., 2025b). These events are increasingly recognized as
59 critical modulators of ozone variability through their impacts on photochemistry and precursor emissions
60 (Gao et al., 2023; Pu et al., 2017; Wang et al., 2022a).

61 Quantifying the respective roles of anthropogenic emissions and meteorological variability in
62 driving ozone trends is therefore essential for evaluating the effectiveness of clean air policies (Li et al.,
63 2019a; Liu et al., 2023). Previous studies have reported rapid increases in surface ozone concentrations

64 in urban cluster regions in China – such as the Beijing–Tianjin–Hebei (BTH) and Yangtze River Delta
65 (YRD) – during the first phase (2013 – 2017), with increases of approximately 28% and 18%,
66 respectively (Chen et al., 2020; Li et al., 2019a; Liu et al., 2023). In contrast, a modest decline in ozone
67 levels was observed during 2018 – 2020, largely attributed to emission reductions (Li et al., 2021; Liu
68 and Wang, 2020b; Wang et al., 2024b; Wang et al., 2023a). However, since 2021, observations indicate
69 a renewed increase in ozone concentrations (Fig. S1). These fluctuations suggest oscillating trends over
70 the past decade, the drivers of which remain poorly constrained.

71 Two main approaches have been applied to attribute air pollution trends: chemical transport models
72 (CTMs) (Li et al., 2021; Liu et al., 2023; Liu and Wang, 2020a) and data-driven statistical frameworks
73 (Li et al., 2019a; Li et al., 2019b; Li et al., 2020). The CTMs simulate atmospheric composition based
74 on emission inventories, meteorological fields, and chemical mechanisms (Ivatt et al., 2022; Liu and Shi,
75 2021; Liu et al., 2023; Ye et al., 2024). They allow attribution of trend components to emissions or
76 meteorology, and can also resolve sector-specific impacts. However, these models face challenges,
77 including uncertainties and temporal lags in emission inventories. Statistical models, on the other hand,
78 rely on observational datasets and predictor-response relationships without requiring explicit emissions
79 or chemical schemes (Li et al., 2019a; Li et al., 2019b; Li et al., 2020; Zhai et al., 2019). With the growing
80 availability of atmospheric big data, statistical and machine learning models have emerged as useful tools
81 for trend attribution (Dai et al., 2023; Grange et al., 2018; Vu et al., 2019; Zhang et al., 2025; Zheng et
82 al., 2023). For instance, Grange et al. (2018) developed a random forest-based framework to isolate
83 meteorological influences on particulate matter. Similarly, Wang et al. (2023) used an enhanced **extreme**
84 **gradient boosting** (XGBoost) model to analyze spatial and temporal ozone patterns in China from 2010
85 to 2021, confirming that emission reductions played a key role in recent declines. Other recent efforts
86 have extended statistical models to long-term assessments of air pollution drivers under climate change
87 (Wang et al., 2022b).

88 Here, we developed a machine learning-based framework – fixed emission approximation (FEA) –
89 to quantify the respective roles of anthropogenic emissions and meteorological conditions in shaping
90 summertime surface ozone trends in China. Applying the FEA to nationwide observational data from
91 2013 to 2023, we identified three phases of ozone evolution corresponding to the major clean air actions
92 and policy transitions. We further analyzed short-term ozone anomalies associated with extreme weather

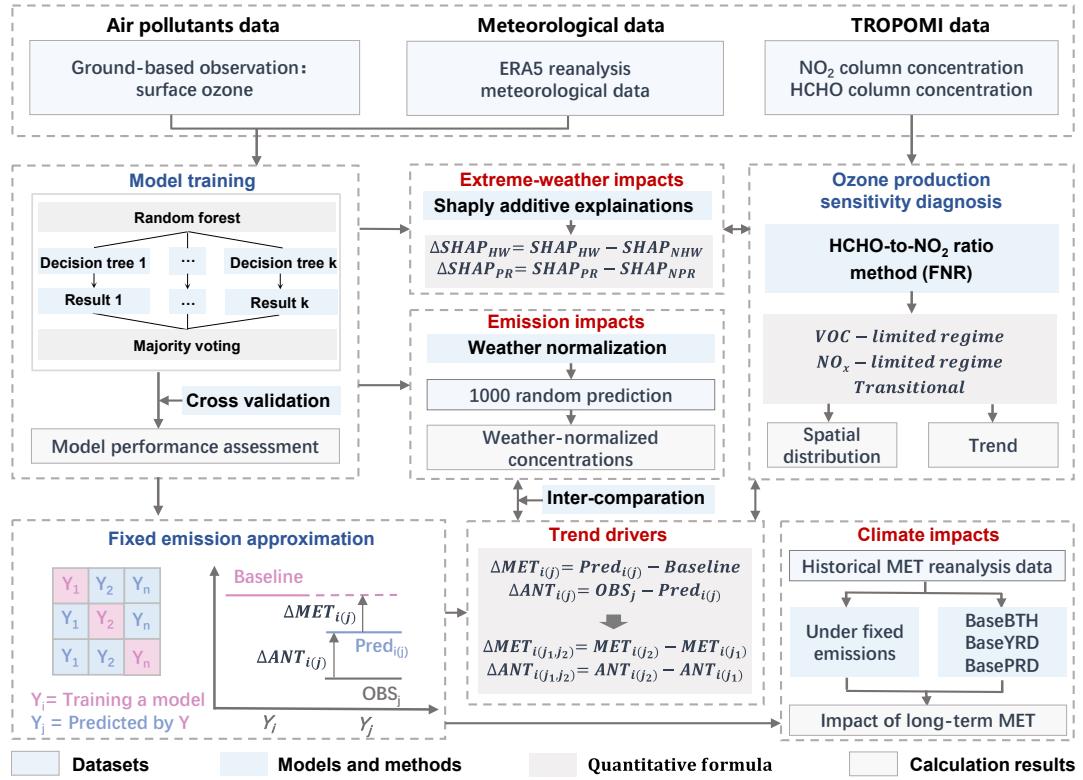
93 events, such as the 2022 heatwave and seasonal rainfall. To characterize ozone production regimes, we
94 integrated satellite-derived formaldehyde-to-nitrogen dioxide (HCHO/NO₂, FNR) ratios from the
95 tropospheric monitoring instrument (TROPOMI), revealing spatiotemporal shifts in ozone formation
96 sensitivity across China. Finally, we extend our FEA analysis to evaluate climate-driven ozone trends
97 from 1970 to 2023, using historical meteorological reanalysis data. Collectively, these analyses provide
98 an integrated understanding of how anthropogenic and climatic factors jointly shape surface ozone
99 dynamics under a warming climate.

100 **2 Data and methods**

101 **2.1 Data sources and methodology overview**

102 Figure 1 provides an overview of the data analysis and methodological framework employed in this
103 study. We first integrated multi-dimensional datasets, including hourly surface air pollutant
104 concentrations, meteorological reanalysis fields, and satellite remote sensing data. Hourly surface
105 observations of ozone, nitrogen dioxide (NO₂), carbon monoxide (CO), and fine particulate matter (PM_{2.5})
106 were accessed from the National Environmental Monitoring Center of China through the open website
107 <https://air.cnemc.cn:18007/> (last accessed: May 20, 2024). Hourly meteorological data with a spatial
108 resolution of 0.25° × 0.25° were sourced from the ERA5 reanalysis dataset provided by the European
109 Centre for Medium-Range Weather Forecasts (ECMWF) and are available for download at
110 <https://cds.climate.copernicus.eu> (last accessed: March 20, 2025). Detailed variables are listed in Table
111 S1. The time variables – hour (hour of day) and month (month of year) – are used as emission surrogates
112 to capture regular diurnal and seasonal variations in anthropogenic activity. A similar strategy is widely
113 applied in previous studies about long-term trends in air pollutants (e.g., Grange et al., 2018; Vu et al.,
114 2019) to separate short-term cyclical emission variability from long-term trends. For 2013 – 2014, the
115 surface MDA8 ozone data were obtained from the Tracking Air Pollution in China (TAP) dataset (Geng
116 et al., 2021), which can be downloaded from <http://tapdata.org> (last accessed: May 20, 2024). The
117 TROPOMI on the Sentinel-5P satellite provides global continuous observation data for two indicators of
118 ozone precursors: NO₂ and formaldehyde (HCHO) column concentrations (Lamsal et al., 2014; Shen et
119 al., 2019). The FNR was used as a proxy for VOC/NO_x reactivity and as a diagnostic indicator of ozone

120 formation sensitivity (Sillman, 1995), to explain and verify the impact of extreme weather and
 121 anthropogenic emissions on ozone. Details of the ozone sensitivity diagnostic method are provided in
 122 Text S1.



139 levels. These temporal emission surrogates, including month and hour, represent short-term regular
140 emission patterns (e.g., diurnal cycles), thereby enabling the model to isolate the long-term emission-
141 driven component of ozone changes (Grange et al., 2018; Meng et al., 2025; Shi et al., 2021; Vu et al.,
142 2019). The meteorological parameters include 18 distinct variables at different altitudes (see Table S1).
143 It should be noteworthy that surface air pollutant observations for each city represent multi-site averages
144 rather than data from a single monitoring station, which reduces the influence of local representativeness
145 errors. The meteorological data are obtained from the nearest grid cell corresponding to each city,
146 ensuring spatial consistency between the pollutant and meteorological datasets. This approach was
147 similar to the methodologies widely adopted in previous studies (Shi et al., 2021; Wang et al., 2025; Yao
148 et al., 2024; Zheng et al., 2023). Our modeling strategy involves building and predicting models for
149 individual cities and for each year from 2015 to 2023, which helps in minimizing the uncertainty caused
150 by surface heterogeneity. Due to the lack of available observational data for many cities in 2013 and
151 2014, we did not develop models for these two years. In our approach, 80% of the dataset is used for
152 model training, while the remaining 20% is reserved for testing. We perform ten-fold cross-validation
153 and assess model performance using seven statistical metrics, as listed in Table S2.

154 Following the construction of the machine learning models for individual cities and years, we
155 introduce the FEA approach. The key principle of FEA is the assumption that the total emissions of ozone
156 precursors remain unchanged from the baseline year. Specifically, we establish hourly-resolution models
157 for the baseline year (i) during the summer season (June to August) as a reference for anthropogenic
158 emissions, represented by the pink solid line in Fig. 1. These models are then applied to predict ozone
159 concentrations under the meteorological conditions of the prediction year (j), while holding the emission
160 levels constant at those of the baseline year (i), as shown by the blue solid line in Fig. 1. The difference
161 between the predicted values ($Pred_i$) and the observed values (OBS_i) for the baseline year (i) represents
162 the model residuals (RES_i), as shown in Eq. (1). The difference in observed MDA8 ozone concentrations
163 between baseline year i and prediction year j is driven by the differences in meteorological conditions
164 ($MET_{i(j)}$) and anthropogenic emission controls ($ANT_{i(j)}$) (Eq. 2). The prediction result $Pred_{i(j)}$ obtained
165 by applying the model trained with data from year i to the meteorological conditions of year j , the
166 difference between $Pred_{i(j)}$ and Baseline ($Pred_i$) is driven by $MET_{i(j)}$, while the difference between
167 $Pred_{i(j)}$ and the observed levels in year j (OBS_j), minus the RES_i , yields the ozone variation driven by

168 ($ANT_{i(j)}$). Therefore, $MET_{i(j)}$ and $ANT_{i(j)}$ can be quantified and calculated using Eqs. (3-4).

169
$$OBS_i = Pred_i + RES_i, \quad (1)$$

170
$$OBS_{i(j)} = MET_{i(j)} + ANT_{i(j)}, \quad (2)$$

171
$$MET_{i(j)} = Pred_{i(j)} - Pred_i, \quad (3)$$

172
$$ANT_{i(j)} = OBS_j - Pred_{i(j)} - RES_i, \quad (4)$$

173 The difference in observed MDA8 ozone concentrations between two different prediction years (j_1 ,
174 j_2) is driven by the differences in meteorological conditions ($\Delta MET_{i(j_1,j_2)}$) and anthropogenic emission
175 controls ($\Delta ANT_{i(j_1,j_2)}$) (Eq. 5). The term $\Delta MET_{i(j_1,j_2)}$ represents the changes in meteorological
176 conditions and can be calculated by the difference between the predicted values, $Pred_{i(j_1)}$ and
177 $Pred_{i(j_2)}$, for the corresponding years (Eq. 6). Similarly, the value of $\Delta ANT_{i(j_1,j_2)}$, representing the
178 change in anthropogenic emissions between the two years j_1 and j_2 , can be therefore calculated using Eq.
179 (7). By performing these calculations, we can isolate and quantify the contributions of meteorological
180 conditions and anthropogenic emission controls to the observed ozone trends. We used a cross-matrix
181 research method to assess the uncertainty of FEA, with specific formulas available in Supporting Method
182 S2.

183
$$\Delta OBS_{(j_1,j_2)} = \Delta MET_{i(j_1,j_2)} + \Delta ANT_{i(j_1,j_2)}, \quad (5)$$

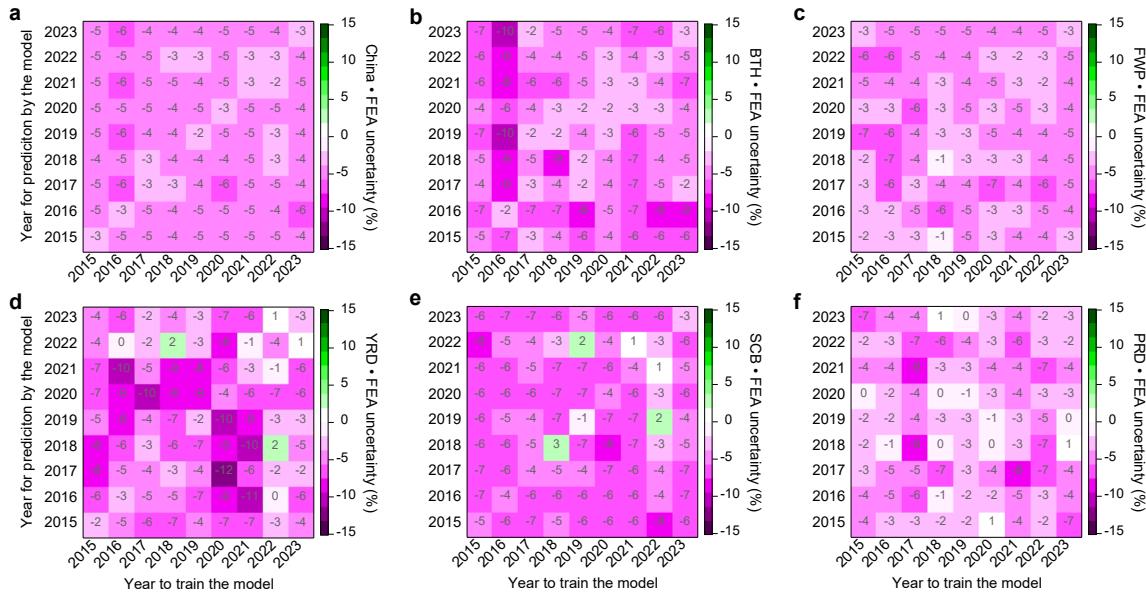
184
$$\Delta MET_{i(j_1,j_2)} = Pred_{i(j_2)} - Pred_{i(j_1)}, \quad (6)$$

185
$$\Delta ANT_{i(j_1,j_2)} = ANT_{i(j_2)} - ANT_{i(j_1)} = (OBS_{j_2} - Pred_{i(j_2)} - RES_i) - (OBS_{j_1} - Pred_{i(j_1)} - RES_i)$$

186
$$= (OBS_{j_2} - OBS_{j_1}) - (Pred_{i(j_2)} - Pred_{i(j_1)}), \quad (7)$$

187 Model performance was first evaluated through ten-fold cross-validation for the BTH region,
188 revealing high predictive skill between observed and predicted MDA8 ozone levels during 2015-2023
189 (Fig. S2). The index of agreement (IOA) ranged from 0.96 to 0.97, with correlation coefficients (R)
190 between 0.93 and 0.95. Root mean square errors (RMSE) and normalized mean bias (NMB) varied from
191 16.9 to 21.9 $\mu\text{g m}^{-3}$ and 8 to 25%, respectively, indicating high model accuracy. Nationally, the model
192 yielded R values of 0.88–0.91 and IOA of 0.93–0.95, with errors remaining within acceptable ranges
193 (Tables S3–S8). To assess uncertainty stemming from interannual model training variability, we applied
194 a matrix-based resampling approach (see Text S2). As shown in Fig. 2, the relative difference in residuals

195 ranged from -9% to 3%, and remained within $\pm 12\%$ for all regions – supporting the robustness of the
 196 FEA method. We found that the model with the added time variables exhibited significantly smaller
 197 uncertainty compared to the model without it (Fig. S3).



198
 199 **Figure 2. Uncertainty assessment of the FEA method.** The uncertainty for the FEA method is calculated using the
 200 approach described in Text S2. The diagonal line in each sub-panel represents the changes in the residuals of the
 201 models.

202 **2.3 Weather normalization analysis**

203 To compare the FEA method with other commonly used statistical approaches, we also applied the
 204 widely adopted meteorological normalization technique based on the RF algorithm. This approach
 205 constructs a regression model that relates air pollutant concentrations to meteorological parameters and
 206 emission surrogate indicators (i.e., time variables such as unix time, day of year, day of month, and hour
 207 of day) (Grange et al., 2018; Vu et al., 2019). Once the model is trained, pollutant concentrations are
 208 predicted by randomly resampling meteorological variables from long-term historical meteorological
 209 datasets, thereby generating a new ensemble of predictions (Vu et al., 2019). These predictions are made
 210 under consistent meteorological conditions, enabling the isolation of meteorological influences from
 211 anthropogenic emission effects on air pollutant trends. The resulting weather-normalized pollutant
 212 concentrations (Fig. 1) represent the levels expected under average meteorological conditions, thus
 213 reflecting the impact of emission changes alone. This approach, first proposed by Grange et al. (2018),
 214 has been widely applied in the long-term attribution of air pollution trends and in assessing short-term
 215 emission reduction effects (Shi et al., 2021; Vu et al., 2019). In this study, the meteorological

216 normalization follows this established framework, with meteorological variables randomly sampled from
217 the long-term dataset spanning 1970-2023. Each normalization process involves 1,000 iterations, and the
218 arithmetic mean of these iterations' simulated values is adopted as the final normalized result. The
219 alignment between FEA-based and weather-normalized trends (Fig. S4) affirms the robustness of the
220 FEA framework.

221 **2.3 Quantification of extreme weather-driven changes in ozone**

222 An unprecedented and prolonged heat wave in the summer of 2022 struck central and eastern China,
223 with the most severely affected area being the YRD (Wang et al., 2023b; Zhang et al., 2023), identified
224 as the longest-lasting and most intense heat wave since at least 1961 (Mallapaty, 2022). The Yangtze-
225 Huaihe region, where the prolonged rainfall occurs, is also a region of frequent sustained extreme
226 precipitation events (Yin et al., 2020). The opening of the “rainy season” is marked in late June with the
227 northward push of the East Asian summer winds and the first northward jump of the subtropical high-
228 pressure ridgeline in the western Pacific Ocean, and generally lasts until mid-July.

229 We employed the SHapley Additive exPlanations (SHAP) method (Lundberg et al., 2020) to
230 elucidate the potential impacts of all input features k on the predictions of the RF model. SHAP assigns
231 an importance score to each feature, revealing their respective contributions to model predictions.
232 Positive values indicate a beneficial influence on predictions, while negative values denote adverse
233 effects. The extreme heatwave events in 2022 (HW) and prolonged rainfall (PR) provide unique and
234 realistic atmospheric environments for us to explore the effects of typical weather extremes on ozone.
235 The PR period is defined as June 15 to July 15 each year, with the remaining period from June to August
236 categorized as the non-prolonged rain period (NPR). To isolate the effects of the rainy season, we defined
237 the HW event in 2022 as occurring from July 16 to August 31, while the corresponding periods in other
238 years were considered non-heatwave (NHW) periods. We first calculated the SHAP values for input
239 features during PR and NPR, as well as HW and NHW periods. The relative changes in SHAP values
240 ($\Delta SHAP$) between these conditions were used to assess the responses of MDA8 ozone to the rainy season
241 and the 2022 heatwave, as per the following Eqs:

$$242 \Delta SHAP_k = SHAP_{PR} - SHAP_{NPR} \quad (8)$$

$$243 \Delta SHAP_k = SHAP_{HW} - SHAP_{NHW} \quad (9)$$

244 **2.5 FEA-based assessment of climate change impacts on ozone**

245 To evaluate the long-term impact of climate change on surface ozone concentrations across
246 China from 1970 to 2023, we extended the framework of our machine learning-based FEA method.
247 The core idea of this analysis is to isolate the influence of long-term meteorological variability on
248 ozone while assuming fixed anthropogenic emissions. Given the availability of relatively complete
249 and continuous hourly ozone observations and meteorological data from 2015 to 2023, this period
250 was used to construct nine emission baseline scenarios. Following the modeling protocol described in
251 Section 2.2, nine independent random forest models were trained for each city and scenario, with each
252 year from 2015 to 2023 serving as an emissions reference. Model inputs included hourly ozone
253 observations, key meteorological predictors, and time-related variables (hour of day and month of year).
254 The trained models were then applied to historical meteorological reanalysis data from 1970 to 2023 to
255 simulate ozone trends under fixed emissions (Fig. 1), yielding nine independent ozone trajectories, each
256 reflecting the influence of long-term meteorological variability under a different fixed-emissions
257 assumption. While the choice of emission baseline may affect the absolute magnitude of simulated ozone,
258 it does not alter the primary objective: assessing the sensitivity of surface ozone to meteorological drivers
259 over multidecadal timescales (Lecœur et al., 2014; Leung et al., 2018; Wang et al., 2022b). This approach
260 could capture the climate-induced ozone signal while adopting the common assumption that emissions
261 are not themselves influenced by climate change – a simplification consistent with prior attribution
262 studies (Dang and Liao, 2019; Leung et al., 2018; Shen et al., 2017; Wang et al., 2022b). For comparison,
263 we also estimated the impact of anthropogenic emission changes on ozone concentrations during 2015–
264 2023 using the same FEA methodology and the complete hourly dataset for model training. This dual-
265 track analysis enables a clear distinction between the impacts of climate variability and emission
266 mitigation on observed ozone trends.

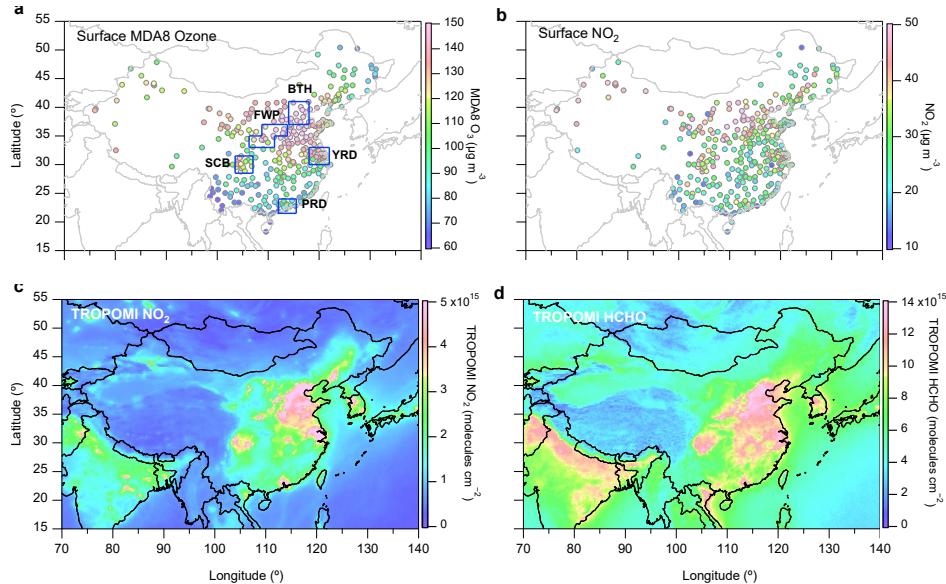
267 To examine the sensitivity of urban ozone pollution to climate variability under different potential
268 atmospheric conditions (e.g., oxidation capacity) and its possible evolution under global warming, we
269 defined three representative regional scenarios based on typical ozone pollution characteristics in China
270 (Fig. 3a): a high-pollution scenario for BTH (BaseBTH), a moderate-pollution scenario for YRD
271 (BaseYRD), and a low-pollution scenario for Pearl River Delta (PRD) (BasePRD). These scenarios allow
272 assessment of ozone trends and climate impacts under fixed emissions across three distinct atmospheric

273 conditions.

274 **3 Results and Discussion**

275 **3.1 Spatiotemporal evolution of summertime ozone (2013–2023)**

276 Figure 3 presents the spatial distribution of the average summertime (2018–2023) maximum daily
277 8-hour average (MDA8) ozone, surface NO₂, and TROPOMI NO₂, HCHO column concentrations across
278 China, along with the locations of the country's five major city clusters: Beijing-Tianjin-Hebei (BTH),
279 Fenwei Plain (FWP), Yangtze River Delta (YRD), Sichuan Basin (SCB), and Pearl River Delta (PRD).
280 Across these five major city clusters, the average summer ozone concentrations ranged from 88.9 to
281 161.3 $\mu\text{g m}^{-3}$ – substantially exceeding the 43.0 $\mu\text{g m}^{-3}$ threshold associated with ecosystem productivity
282 loss (Gong et al., 2021) and the World Health Organization (WHO, 2021)-recommended peak seasonal
283 average of 60 $\mu\text{g m}^{-3}$. TROPOMI satellite observations of NO₂ column concentration show notably
284 elevated concentrations over the five major city clusters, particularly in the BTH, YRD, and FWP, which
285 align with surface NO₂ distribution patterns and confirm the scale of anthropogenic NO_x emissions in
286 these regions (Zheng et al., 2021). TROPOMI satellite observations of HCHO column concentrations
287 similarly reveal these city clusters as hotspots for VOC emissions (Fig. 3d). These concurrent high levels
288 of NO₂ and HCHO suggest a strong photochemical ozone pollution potential, as the abundant precursors
289 in these urban clusters could drive substantial ozone production during the summer months. This
290 highlights the significant risks posed by summertime ozone in China's most urbanized and industrialized
291 regions, with implications for both human health and ecosystem.

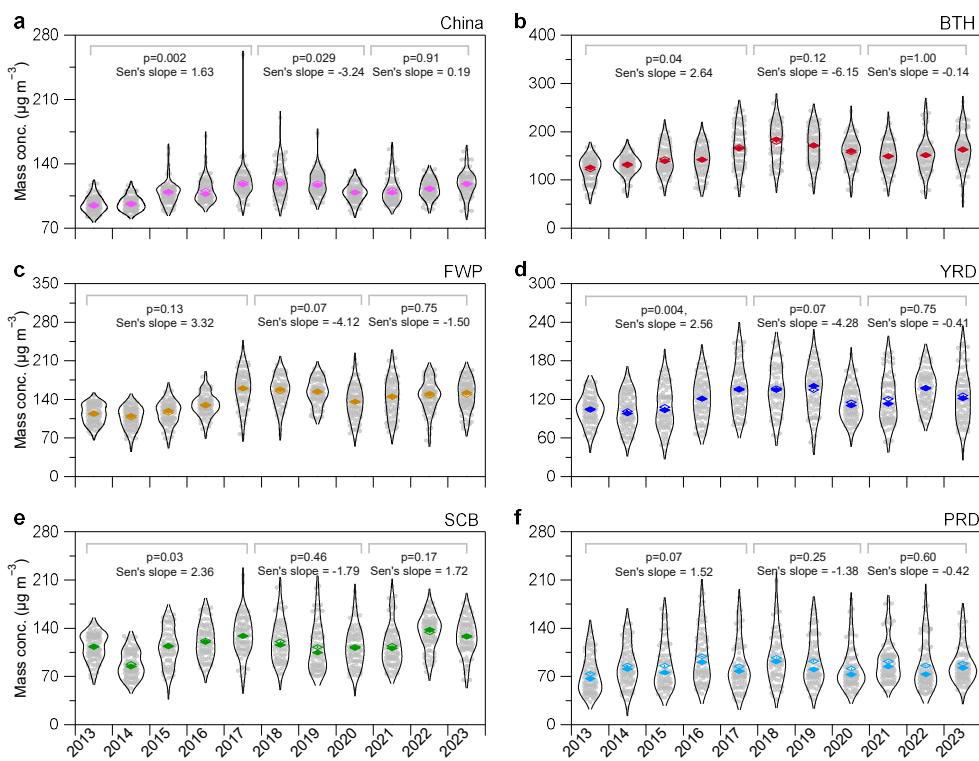


292
293 **Figure 3. Spatial distribution of summertime MDA8 ozone, surface NO₂, and TROPOMI NO₂, HCHO across**
294 **major city clusters in China.** The panels represent the average MDA8 ozone, surface NO₂, and TROPOMI NO₂,
295 HCHO column concentrations for 354 cities in China during the summertime (June–August) from 2018 to 2023.
296 The corresponding five regions includes BTH (37°–41°N, 114°–118°E); YRD (30°–33°N, 118.2°–122°E); SCB
297 (28.5°–31.5°N, 103.5°–107°E); PRD (21.5°–24°N, 112°–115.5°E) and FWP (106.25–111.25°E, 33–35°N, and
298 108.75–113.75°E, 35–37°N).

299

300 Figure 4 presents the interannual variations in MDA8 ozone concentrations during summertime
301 across China, with a focus on five key urban clusters. During the first phase (2013–2017), the average
302 nationwide MDA8 ozone increased significantly ($p < 0.01$), rising from 95.5 to $118.0 \mu\text{g m}^{-3}$. This growth
303 was especially pronounced in the BTH and FWP regions, where concentrations increased by
304 approximately 38% and 41%, respectively. In contrast, ozone increases were more modest in the YRD
305 (~11%), SCB (~15%), and PRD (~16%) regions, respectively. These results were consistent with the
306 previous studies (Li et al., 2021; Liu and Wang, 2020a, 2020b; Wang et al., 2023a). In the second phase,
307 corresponding to the implementation of more stringent emission controls on NO_x and VOCs emissions
308 (Geng et al., 2024; Liu et al., 2023), a moderate national decrease in MDA8 ozone was observed, with
309 concentrations declining to $109.0 \mu\text{g m}^{-3}$ from 2017 to 2020. The declines during this period were most
310 notable in FWP (~16%) and YRD (~15%), while BTH (~6%), SCB (~11%), and PRD (~4%) also showed
311 reductions compared to their concentration peaks observed in 2017. However, the MDA8 ozone
312 rebounded, reaching $118.4 \mu\text{g m}^{-3}$ in 2023 – comparable to its 2017 peak – with a particularly sharp
313 increase during the summer of 2022. From 2021 to 2023, MDA8 ozone concentrations rose by $2.8 \mu\text{g m}^{-3}$
314 in BTH, $3.1 \mu\text{g m}^{-3}$ in FWP, $16.1 \mu\text{g m}^{-3}$ in YRD, and $18.5 \mu\text{g m}^{-3}$ in SCB, respectively.

315 Figure S1 further illustrates the spatiotemporal evolution of summertime MDA8 ozone in China
 316 from 2013 to 2023. On average, approximately 68% of cities exceeded the WHO air quality guideline of
 317 $100.0 \mu\text{g m}^{-3}$ for the MDA8 ozone. Elevated ozone levels were primarily observed in densely populated
 318 and economically developed regions. Spatially, ozone hotspot regions expanded between 2013 and 2017
 319 (Fig. S1 a-e), followed by contraction during 2018-2020 (Fig. S1 f-i). However, this progress stalled in
 320 2021. A sharp reversal was observed in 2022, with widespread increases in MDA8 ozone (Fig. S1 k).
 321 These changes could be closely linked to emission control measures and meteorological conditions,
 322 which will be further discussed in Sections 3.2 and 3.3.



323
 324 **Figure 4. Interannual trends of summertime MDA8 ozone across China (2013–2023).** Panel (a) illustrates the
 325 seasonal variations of MDA8 ozone during the summer months (June, July, and August) across 354 cities nationwide.
 326 Panels (b-f) show the average trend across five key regions in China: BTH, FWP, YRD, SCB, and PRD. The summer
 327 months are defined according to meteorological seasonality, encompassing June, July, and August. In the violin plots,
 328 hollow diamond markers denote the mean, while solid diamond markers represent the median. The Mann-Kendall
 329 test and Sen's slope estimator were employed to assess the statistical significance and rate of change in the monthly
 330 average MDA8 ozone concentrations. The *p* value represents the significance level from the Mann-Kendall test,
 331 which is used to determine the statistical significance of the trend in the data.

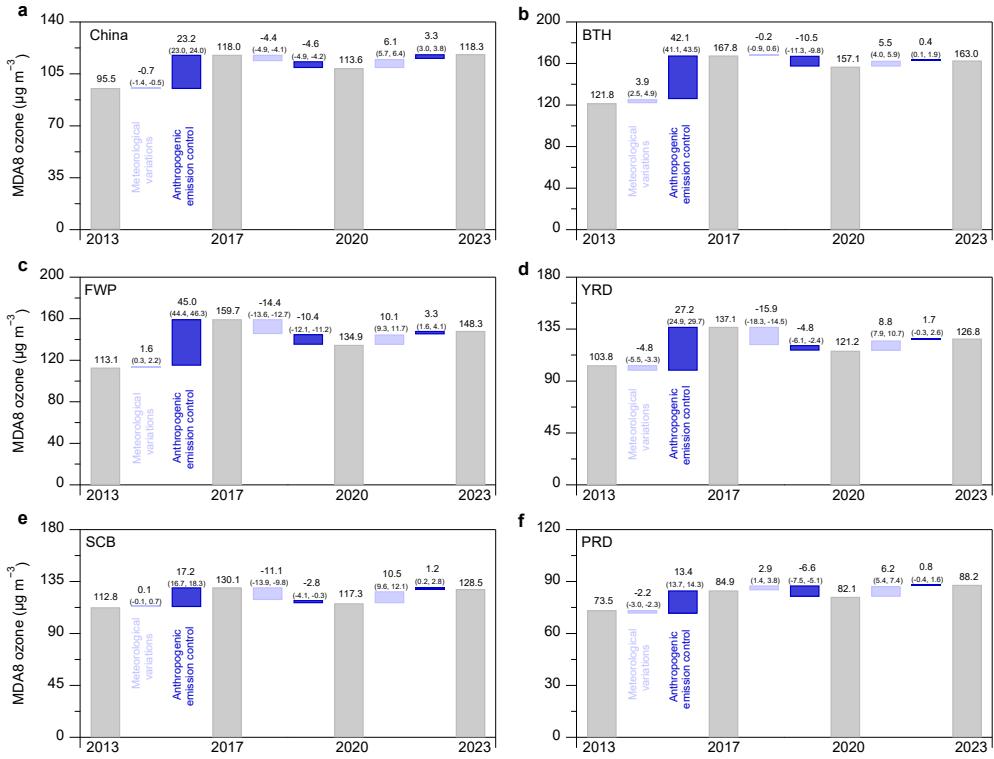
332

333 3.2 Anthropogenic emission drivers of ozone trends

334 To disentangle the relative impacts of anthropogenic emissions and meteorological variability on

335 observed ozone trends, we applied the machine learning-based FEA framework described in Section 2.2.
336 As illustrated in Fig. 5, anthropogenic emissions were the dominant driver of ozone increases during
337 2013–2017, contributing an average rise of approximately $23.2 \pm 1.1 \text{ } \mu\text{g m}^{-3}$ nationwide. The most
338 pronounced increases occurred in the FWP and BTH ($45.0 \pm 2.0 \text{ } \mu\text{g m}^{-3}$ and $42.1 \pm 2.0 \text{ } \mu\text{g m}^{-3}$,
339 respectively), whereas the PRD exhibited a relatively modest enhancement ($13.4 \pm 1.6 \text{ } \mu\text{g m}^{-3}$), reflecting
340 its predominantly NO_x-limited photochemical regime versus VOC-limited regimes in other regions (Ren
341 et al., 2022). As shown in Fig. S5, the precursor gases NO₂ and CO exhibited regionally distinct
342 decreasing trends, partially explaining the spatial heterogeneity of ozone changes. During the second
343 phase, the MDA8 ozone decreased by $10.5 \pm 2.0 \text{ } \mu\text{g m}^{-3}$ in BTH and $10.4 \pm 3.0 \text{ } \mu\text{g m}^{-3}$ in FWP, with
344 smaller declines in YRD ($-4.8 \pm 3.8 \text{ } \mu\text{g m}^{-3}$), SCB ($-2.8 \pm 2.4 \text{ } \mu\text{g m}^{-3}$), and PRD ($-6.6 \pm 1.4 \text{ } \mu\text{g m}^{-3}$)
345 between 2017 and 2020 (Fig. 5). These trends are broadly consistent with those derived using
346 independent statistical approaches (Wang et al., 2023).

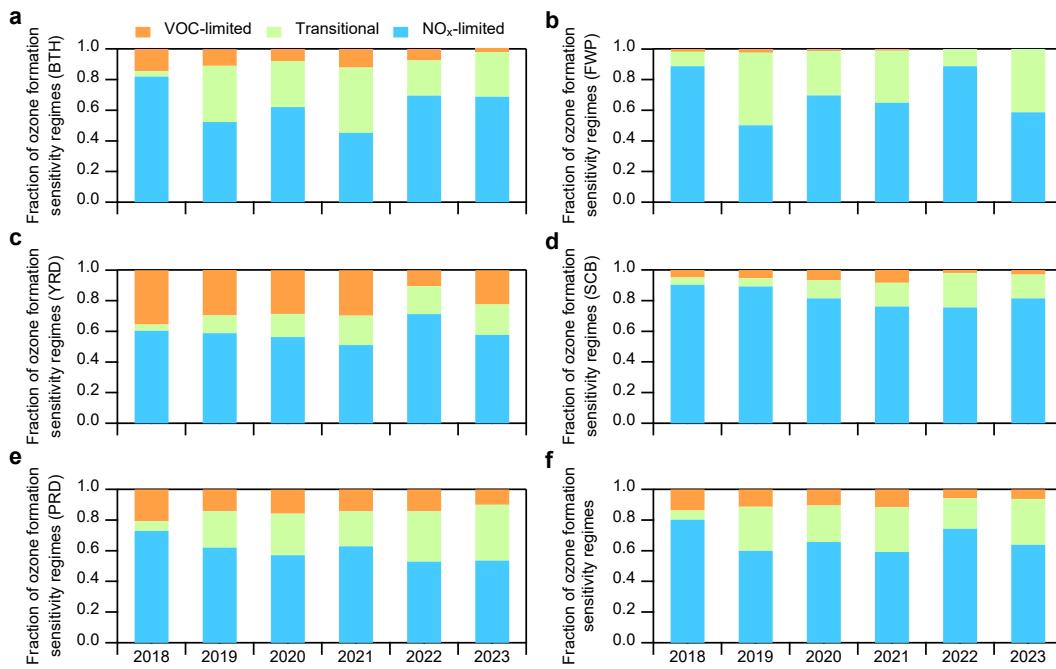
347 The COVID-19 pandemic (January–April 2020) introduced an unprecedented perturbation to
348 anthropogenic activity, leading to sharp declines in industrial production, energy consumption, and
349 transportation (Shi and Brasseur, 2020; Zheng et al., 2021). National emissions of SO₂, NO_x, PM_{2.5}, and
350 VOCs were estimated to have decreased by 0.37 Tg (12%), 0.87 Tg (13%), 0.25 Tg (10%), and 1.07 Tg
351 (12%), respectively, relative to the same period in 2019 (Geng et al., 2024). Despite these reductions,
352 MDA8 ozone concentrations increased by 1.7–2.3 $\mu\text{g m}^{-3}$ across BTH, FWP, YRD, and SCB, while a
353 slight decrease occurred in PRD (Fig. S6). Overall, ~79% of monitored cities experienced ozone
354 increases, with a national mean enhancement of $2.1 \pm 1.3 \text{ } \mu\text{g m}^{-3}$ (Fig. S7). In the post-pandemic period
355 (2020 – 2023), concentrations of NO₂, CO, and PM_{2.5} stabilized or declined more gradually (Fig. S5),
356 and the contribution of anthropogenic emissions to ozone variability weakened considerably (Fig. S8).
357 Regionally, emission-driven changes ranged from -1.2 to $+2.6 \text{ } \mu\text{g m}^{-3}$ in BTH, -1.6 to $+4.0 \text{ } \mu\text{g m}^{-3}$ in
358 FWP, -4.7 to $+7.4 \text{ } \mu\text{g m}^{-3}$ in YRD, -3.6 to $+3.0 \text{ } \mu\text{g m}^{-3}$ in SCB, and -3.8 to $+7.7 \text{ } \mu\text{g m}^{-3}$ in PRD. These
359 results indicate that while emission control policies initially produced substantial benefits, their
360 effectiveness has gradually diminished, suggesting that ozone responses to further emission reductions
361 may have reached a saturation point.



362 **Figure 5. Anthropogenic and meteorological drivers of ozone trends from 2013 to 2023.** Changes in summertime
 363 MDA8 ozone concentrations were decomposed into contributions from anthropogenic emissions and meteorological
 364 variability using the FEA framework. Results reflect ensemble estimates based on multiple baseline years (2015–
 365 2023) for emissions. Boxplots indicate the interquartile range, with values in parentheses denoting the 25th and 75th
 366 percentiles across all baseline scenarios.

368
 369 Satellite retrievals of tropospheric NO₂ and HCHO from TROPOMI (Figs. S9 – S10) further reveal
 370 evolving ozone production chemistry. NO₂ columns exhibited strong east – west gradients, with eastern
 371 China maintaining levels five times higher than the west. Between 2018 and 2023, NO₂ columns over
 372 the North China Plain (NCP) declined from 4.13×10^{15} to 3.85×10^{15} molecules cm⁻², while HCHO
 373 remained stable until 2021, followed by a sharp increase in 2022. **The spatial pattern of temperature**
 374 **anomalies between heatwave (HW) and non-heatwave (NHW) periods (Fig. S11)** reveals strong positive
 375 differences in the YRD and SCB, consistent with enhanced biogenic and anthropogenic VOC emissions
 376 under extreme heat (Qin et al., 2025; Tao et al., 2024). By 2023, HCHO concentrations had returned to
 377 pre-heatwave levels. To diagnose the evolving chemical sensitivity of ozone production, we examined
 378 the spatiotemporal evolution of the HCHO/NO₂ ratio (Text S1). Figure S12 shows that this ratio exhibited
 379 regionally distinct transitions from 2018 to 2023, reflecting shifts in photochemical regimes. Figure 6
 380 summarizes the relative contributions of VOC-limited, NO_x-limited, and transitional regimes across the
 381 five key regions. In BTH, NO_x-limited areas accounted for ~82% of the domain in 2018 and remained

382 above 45% thereafter, while VOC-limited regions declined from ~14% to ~2%. In FWP, summer ozone
 383 formation was largely controlled by NO_x-limited and transitional regimes. The YRD underwent a notable
 384 shift from VOC- to NO_x-limited chemistry, with VOC-limited fractions decreasing from ~35% in 2018
 385 to ~22% in 2023, particularly during 2022 when extreme heat amplified VOC emissions and
 386 photochemical activity (Qin et al., 2025; Tao et al., 2024). The SCB region consistently exhibited strong
 387 NO_x limitation (>75%), whereas the PRD showed a gradual expansion of the transitional regime
 388 alongside a modest contraction of VOC-limited areas. These shifts in photochemical sensitivity
 389 correspond well with the ozone decrease observed during Phase II emission reductions. Spatial
 390 distributions of ozone formation sensitivity during the COVID-19 lockdown (Fig. S13) reveal that most
 391 of China was in a transitional regime, with major urban clusters remaining VOC-limited and only limited
 392 areas in southern China being NO_x-limited. This spatial pattern aligns with the observed widespread
 393 ozone increases during the lockdown (Fig. S7). Collectively, these regional transitions reflect the dual
 394 influences of sustained anthropogenic emission controls and short-term climatic perturbations. The
 395 findings highlight that ozone production chemistry in China is becoming increasingly shaped by the
 396 complex interplay between emission reduction efforts and the rising frequency of meteorological
 397 extremes under a warming climate.



398
 399 **Figure 6. Trends in the distributions of ozone production sensitivity regimes.** Fractions of VOC-limited, NO_x-
 400 limited, and transitional ozone sensitivity regimes across five key regions during the summertime (June to August)
 401 from 2018 to 2023, based on the FNR analysis. Panel (f) presents the overall trends for all five regions.

402 **3.3 Meteorological impact on ozone variation**

403 Figure 5 shows the interannual meteorological contributions to summertime MDA8 ozone across
404 different emission-control phases. During Phase I (2013 – 2017), meteorology exerted relatively weak
405 influences on ozone variability, with contributions ranging from -4.8 to $+3.9 \mu\text{g m}^{-3}$ —far smaller than
406 those from anthropogenic emission changes. In contrast, meteorological anomalies became a decisive
407 factor during Phase II (2017–2020), driving substantial ozone reductions. **Ozone decreases attributable**
408 **to meteorology reached $-14.4 \pm 3.0 \mu\text{g m}^{-3}$ in the FWP, $-15.9 \pm 3.8 \mu\text{g m}^{-3}$ in the YRD, and -11.1 ± 2.4**
409 **$\mu\text{g m}^{-3}$ in the SCB, explaining $58 \pm 12\%$, $77 \pm 18\%$, and $80 \pm 17\%$ of the total ozone decline, respectively.**

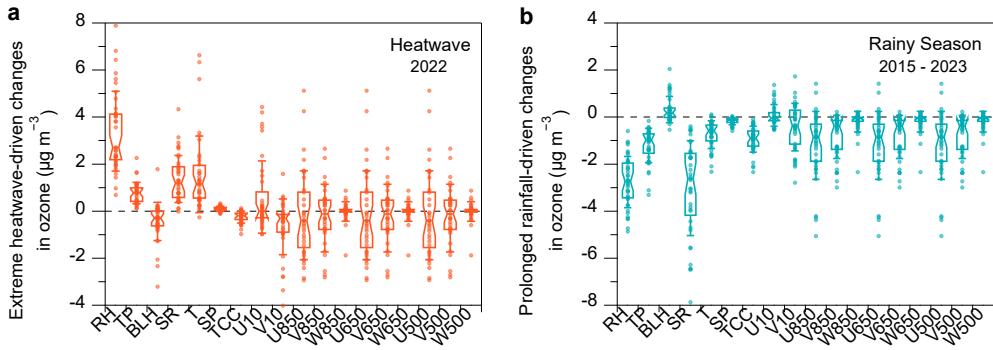
410 A notable shift occurred during Phase III, when the influence of extreme meteorological events
411 increasingly dominated ozone variability. In the summer of 2022, persistent heatwaves across eastern
412 and southern China (Mallapaty, 2022; Wang et al., 2023b) triggered sharp ozone increases of $+20.8 \pm 3.6$
413 $\mu\text{g m}^{-3}$ in the YRD and $+22.1 \pm 3.2 \mu\text{g m}^{-3}$ in the SCB, **reflecting the enhanced photochemical activity**
414 **under high-temperature and intense solar radiation conditions.** The following summer (2023) featured
415 anomalously heavy rainfall, resulting in sharp ozone suppression ($-17.8 \pm 2.3 \mu\text{g m}^{-3}$ in the YRD and $-$
416 $9.7 \pm 3.3 \mu\text{g m}^{-3}$ in the SCB). This reduction coincided with a remarkable increase in precipitation, i.e.,
417 102% in YRD and 35% in SCB (Fig. S14), indicating that rainfall-induced wet scavenging and
418 diminished photolysis substantially curtailed ozone production.

419 To identify the dominant meteorological drivers, we analyzed Gini importance scores derived from
420 the RF model across 18 predictor variables (Fig. S15). Temperature (T) emerged as the most influential
421 predictor in the BTH and FWP regions, while shortwave solar radiation (SR), relative humidity (RH),
422 and 850hPa zonal wind (u850) were most important in the YRD. In the PRD, ozone variability was
423 primarily governed by temperature and transport-related indices, including meridional winds at different
424 altitudes. These findings are consistent with the climatological contrast between northern continental and
425 southern coastal regimes: in northern China, stagnant anticyclonic conditions (Gong and Liao, 2019) and
426 strong solar radiation promote photochemical buildup (Bao et al., 2025), whereas in southern regions,
427 high humidity and convective rainfall tend to suppress ozone by reducing actinic flux and enhancing
428 removal of precursors.

429 Partial dependence analysis (Fig. S16) further illustrates the nonlinear responses of ozone to key
430 meteorological factors (T , RH, SR) for representative cities in each cluster, revealing clear regional

431 contrasts. In Beijing (BTH), ozone concentrations show the strongest positive response to T (Fig. S16a),
432 consistent with the enhancement of reaction kinetics and biogenic VOC emissions under hot conditions.
433 This behavior reflects the thermodynamic coupling between surface heating, boundary-layer expansion,
434 and photochemical production. In Nanjing (YRD), ozone is more sensitive to solar radiation than to
435 temperature (Fig. S16c), highlighting the dominant role of actinic flux in controlling radical production
436 during warm and dry conditions. Yang et al. (2024) similarly reported that high-temperature and low-RH
437 conditions over the NCP and YRD enhance photochemical ozone formation, with chemical production
438 being the dominant process driving ozone buildup during the most polluted months. In the SCB, both T
439 and RH exhibit strong influences, while in the PRD, ozone variability is shaped primarily by T and large-
440 scale circulation patterns associated with subtropical maritime flow and typhoon incursions from the
441 Northwest Pacific (Chen et al., 2024; Wang et al., 2024a; Wang et al., 2022a).

442 To further quantify these relationships, we applied SHAP (SHapley Additive exPlanations) analysis
443 to evaluate the meteorological influence of heatwave (HW) and pluvial (PR) events in the Yangtze-
444 Huaihe region between 2015 and 2023 (Section 2.4). As shown in Fig. S17, HW events were associated
445 with strong positive SHAP values in southeastern coastal areas, especially the YRD and SCB, driven by
446 elevated SR and T. Mean SR during HW periods was substantially higher than during non-HW periods
447 (Fig. S18), increasing photochemical activity through increased radical generation and faster reaction
448 rates. Conversely, PR events produced consistent negative SHAP contributions across all regions (Fig.
449 S19), reflecting the combined effects of reduced photolysis, increased humidity, and efficient wet
450 scavenging on ozone production (He and Carmichael, 1999). A multi-year comparison (Fig. 7) highlights
451 the opposing effects of key meteorological variables – including RH, T, boundary layer height (BLH),
452 total precipitation (TP), and surface pressure (SP) – on MDA8 ozone. The trend in Δ SHAP values under
453 high-humidity conditions from 2015 to 2023 (Fig. S20) further confirms the model's ability to capture
454 the suppressive effects of wet weather conditions on ozone formation.



455 **Figure 7. Meteorological impact on predicted ozone concentrations under heatwave and rainy weather**
456 **conditions.** (a) Differences in SHAP values (Δ SHAP) between heatwave and non-heatwave periods in the Yangtze-
457 Huaihe region during summer 2022. (b) Differences in SHAP values (Δ SHAP) between prolonged rainfall periods
458 and non-prolonged rainfall periods in the same region from 2015 to 2023. Box plots show the distribution of Δ SHAP
459 across cities; the center line indicates the median, boxes denote the interquartile range (25th-75th percentiles), and
460 the whisker line extends to one standard deviation.

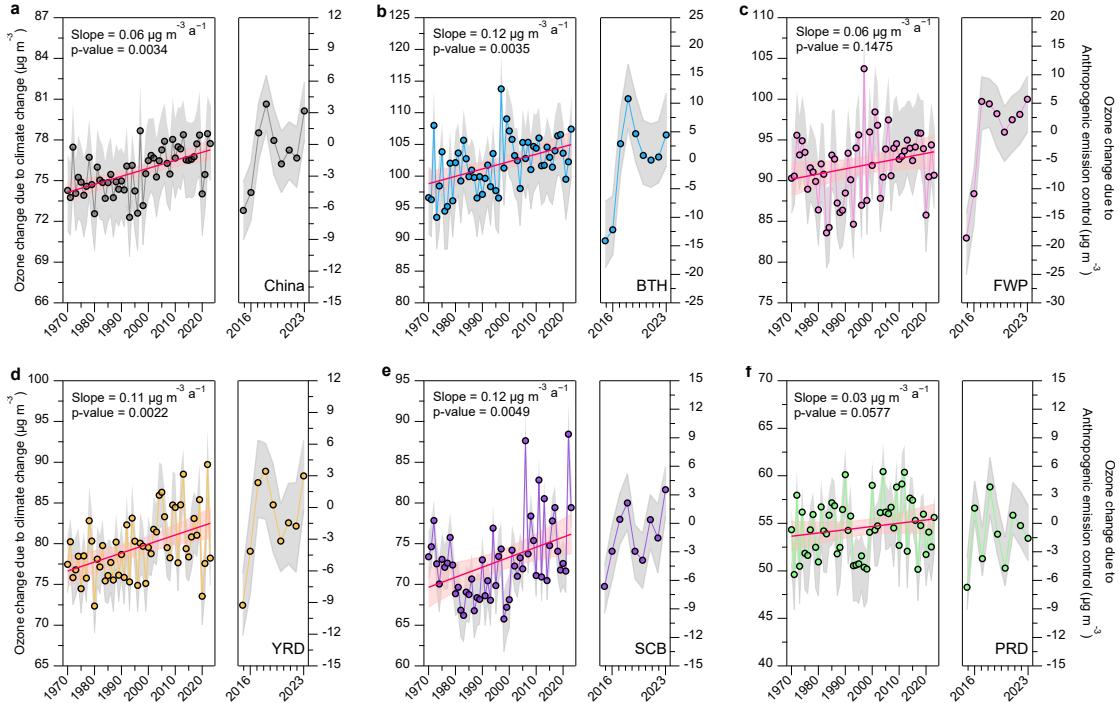
461
462 **3.4 Reshaping distributions of ozone controlled by a warming climate**

463 To assess the long-term influence of climate change on surface ozone concentrations, we applied
464 the FEA framework to simulate summertime ozone trends over the period 1970 – 2023. In this analysis,
465 anthropogenic emissions were held constant at their 2015 – 2023 summertime levels, while interannual
466 variations in meteorological variables were introduced using historical reanalysis data. This design
467 isolates the climate-driven component of ozone trends while assuming that emission trajectories are
468 independent of climate change – a simplification aligned with prior attribution frameworks (Wang et al.,
469 2022b). The impact of anthropogenic emission controls was estimated by comparing observed ozone
470 concentrations with FEA-predicted values during 2015 – 2023, thereby quantifying the residual effect of
471 emissions under fixed meteorology.

472 As shown in Fig. 8, under the 2015-2023 emission levels, climate change has exerted a statistically
473 significant ($p < 0.05$) positive influence on urban summertime ozone concentrations across China,
474 resulting in a nationwide increase of approximately $0.06 \mu\text{g m}^{-3} \text{ yr}^{-1}$ since 1970. All five major urban
475 regions displayed upward trends, with the most pronounced increase observed in the BTH and SCB at
476 $0.12 \mu\text{g m}^{-3} \text{ yr}^{-1}$. Three sensitivity simulations (see Section 2.5 and Fig. S21) confirm this robustness:
477 trend slopes range from $0.11\text{--}0.14 \mu\text{g m}^{-3} \text{ yr}^{-1}$ in BaseBTH, $0.05\text{--}0.10 \mu\text{g m}^{-3} \text{ yr}^{-1}$ in BaseYRD, and
478 $0.03\text{--}0.10 \mu\text{g m}^{-3} \text{ yr}^{-1}$ in BasePRD. Despite regional differences in chemical regimes or pollution levels,
479 the consistent upward tendencies underscore the pervasive climatic amplification of ozone formation.

481 These results emphasize that climate change acts as a systematic driver of ozone growth across diverse
482 atmospheric environments, reinforcing the need to embed climate resilience within emission control
483 strategies. **Spatial correlations between climate-driven ozone increases and temperature changes (Fig.**
484 **S22) further confirm that warming is the dominant contributor to long-term ozone enhancement.** In
485 particular, the correlation coefficients between ozone trends and temperature anomalies reached 0.90
486 (BTH), 0.89 (FWP), 0.72 (YRD), and 0.93 (SCB), indicating a strong temperature dependence of
487 **climate-induced ozone formation in these regions.** The PRD showed a weaker correlation, likely due to
488 its unique subtropical maritime climate and higher humidity and cloud cover, which tend to suppress
489 photochemical ozone production (Yang et al., 2019). The right panel of Fig. 8 depicts summertime ozone
490 trends from 2015 to 2023 under the combined influence of anthropogenic emissions, derived from the
491 FEA method. Ozone concentrations rose across all regions between 2015 and 2018, declined modestly
492 during 2018-2020, and rebounded thereafter in most regions except the PRD.

493 These findings are consistent with future projections that anticipate more frequent high-ozone
494 episodes under continued warming (Li et al., 2023). Recent analyses (Yang et al., 2024) show that the
495 frequency of high-temperature and low-humidity conditions during 2000-2019 was markedly higher than
496 in 1980-1999, suggesting that ozone pollution in both the NCP and YRD has intensified under historical
497 climate change. Indeed, while national emission controls curbed ozone growth after 2018, a post-2020
498 rebound has emerged, implying that the climatic penalty on ozone is beginning to offset emission gains.
499 The extreme 2022 heatwave exemplified this effect, substantially enhancing photochemical activity and
500 shifting ozone sensitivity from VOC-limited to transitional or NO_x-limited regimes. Although reductions
501 in anthropogenic precursor emissions have improved ozone control efficiency, the direct warming-
502 induced enhancement of ozone increasingly interferes with – and in some regions may partially offset –
503 the air-quality gains achieved through emission reduction efforts.



504

505 **Figure 8. Impact of climate change and emission controls on ozone trends.** Left panels show ozone trends
 506 attributable to long-term climate change from 1970 to 2023, simulated under fixed emission scenarios using the FEA
 507 framework. Right panels depict ozone trends from 2015 to 2023, reflecting the impact of anthropogenic emission
 508 controls. Each trajectory represents results based on a distinct emissions baseline year. Shaded grey areas indicate
 509 the interquartile range (25th-75th percentiles), solid red lines denote trend estimates, and light red shading marks the
 510 5th-95th percentile confidence intervals. Statistical significance and trend slopes were assessed using the Mann-
 511 Kendall test. [More details of the sensitivity tests are provided in Fig. S18.](#)

512

4 Conclusions and implications

513 China is confronted with the dual challenges of climate change and ozone pollution. Over the past
 514 decade, summertime ozone concentrations across the country have exhibited complex spatiotemporal
 515 patterns, reflecting the evolving interplay between anthropogenic emissions, meteorological variability,
 516 and large-scale climate dynamics. In this study, we developed and applied a machine learning-based FEA
 517 framework to disentangle and quantify the respective roles of anthropogenic emissions and
 518 meteorological drivers in shaping ozone trends during 2013-2023. With a national-level prediction
 519 uncertainty of approximately 6%, the FEA method provides a computationally efficient and scalable tool
 520 for diagnosing atmospheric variability across large spatial and temporal domains. [Nonetheless, some](#)
 521 [limitations remain. The current implementation did not explicitly resolve grid-scale spatial heterogeneity,](#)
 522 [vegetation, or land-use dynamics, which may influence ozone formation. Moreover, potential](#)

523 sensitivities to spatial resolution warrant further investigation through coupled applications of machine
524 learning and chemical transport models.

525 Our analysis revealed that increased anthropogenic precursor emissions were the dominant driver
526 of the sharp rise in summertime MDA8 ozone concentrations during the first phase (2013–2017),
527 contributing an average increase of $23.2 \pm 1.1 \text{ } \mu\text{g m}^{-3}$. In contrast, during the second phase (2018–2020),
528 enhanced air quality regulations – particularly the synergistic control of NO_x and VOCs – led to
529 measurable reductions in MDA8 ozone, with national-average declines of $4.6 \pm 1.5 \text{ } \mu\text{g m}^{-3}$. These
530 improvements were especially evident in regions such as BTH and FWP, where ozone formation is highly
531 sensitive to VOC levels. However, during the most recent period (2021–2023), the impact of emission
532 reductions diminished considerably, with regional ozone levels either plateauing or rebounding. This
533 stagnation underscores the urgent need for more targeted, region-specific emission control strategies that
534 address the shifting photochemical sensitivity of ozone formation regimes.

535 Using SHAP attribution analysis, we further quantified the influence of meteorological extremes on
536 ozone variability. Record-breaking heatwaves in 2022 enhanced ozone concentrations by up to $+5.8 \text{ } \mu\text{g}$
537 m^{-3} , while prolonged pluvial episodes—particularly during the East Asian monsoon season—suppressed
538 ozone by as much as $-15.2 \text{ } \mu\text{g m}^{-3}$. These results highlight the growing dominance of short-term
539 meteorological extremes in shaping ozone air quality under a warming climate. Complementary satellite-
540 based FNR diagnostics revealed that most urban clusters remain VOC-limited or transitional, except the
541 PRD, which is largely NO_x-limited. The 2022 heatwave induced regime shifts in regions such as the
542 YRD, where intensified VOC emissions and elevated temperatures drove transitions toward NO_x-limited
543 conditions. These findings emphasize the need for dynamic, region-specific assessments of ozone
544 sensitivity to guide effective mitigation strategies.

545 To assess the climate penalty on ozone, we extended the FEA framework to simulate long-term
546 trends from 1970 to 2023 by fixing emissions and allowing meteorological variables to evolve with
547 observed climate trends. Our findings show that climate change has contributed to a significant upward
548 trend in urban summertime ozone, averaging $0.06 \text{ } \mu\text{g m}^{-3} \text{ a}^{-1}$, with particularly strong increases in the
549 BTH and SCB. Correlations between ozone and surface temperature were consistently high ($r = 0.72$ –
550 0.93) in BTH, FWP, YRD, and SCB, suggesting that warming has increasingly offset gains from emission
551 controls in recent years. **Although reductions in anthropogenic precursor emissions have improved ozone**

552 control efficiency, the direct warming-induced enhancement of ozone increasingly interferes with – and
553 in some regions may partially offset – the air-quality gains achieved through emission reduction efforts.
554

555 *Data availability.* Data are provided within the manuscript or supplementary information files.

556

557 *Code availability.* The statistical computing in this study was based on R language software which can
558 be download at <https://www.r-project.org/>.

559

560 *Author contributions.*

561 Y.Z. and X.G. initiated and designed the study. Y.Z. and JF developed the statistical methodology, model
562 calculation, and data analysis. J.F. and Y.Z. prepared the manuscript with contributions from D.H., B.Z.,
563 M.W., J.L., Y.S., H.L., J.W., Y.W., M.C., and X.G..

564

565 *Competing interests.* The authors declare no competing interests.

566

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573

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