



# **Tracking surface ozone responses to clean air interventions under a warming climate in China**

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16 **Abstract.** Surface ozone, a major air pollutant with profound implications for human health, ecosystems,  
17 and climate, shows long-term trends shaped by both anthropogenic and climatic drivers. Here, we  
18 develop a machine learning-based approach – the Fixed Emission Approximation (FEA) – to disentangle  
19 the effects of meteorological variability and anthropogenic emissions on summertime ozone trends in  
20 China. We identify three distinct phases of ozone trends corresponding to clean air actions.  
21 Anthropogenic emissions drove a  $+23.2 \pm 1.1 \mu\text{g m}^{-3}$  increase in summer maximum daily 8-hour average  
22 ozone during 2013–2017, followed by a  $-4.6 \pm 1.5 \mu\text{g m}^{-3}$  decrease during 2018–2020. However, during  
23 2021–2023, extreme meteorological anomalies – including heatwaves and extended monsoon  
24 rainfall – emerged as key drivers of ozone variability. Satellite-derived formaldehyde-to-nitrogen  
25 dioxide ratios reveal widespread urban volatile organic compounds-limited regimes, with a shift toward  
26 nitrogen oxides-limited sensitivity under influence of heatwaves. Finally, we assess ozone trends under  
27 sustained climate warming from 1970 to 2023 based on the FEA framework. The results indicate a  
28 significant climate-driven increase in ozone levels across China's urban agglomerations,  
29 underscoring the amplifying role of climate change in ozone pollution. Together, these findings  
30 highlight the dual influence of anthropogenic and climatic factors on ozone pollution and emphasize  
31 the need for integrated strategies that couple emission mitigation with climate adaptation to  
32 effectively manage ozone risks in a warming world.

33



## 1 Introduction

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

Long-term ozone variability is jointly influenced by anthropogenic emissions and weather conditions as well as regional climate (Hallquist et al., 2016; Li et al., 2019b; Wang et al., 2022b). While emission controls directly regulate precursor abundance, climate change modulates ozone through chemical feedbacks, meteorological dynamics, and biosphere – atmosphere interactions (Ma and Yin, 2021; Xue et al., 2020). Over the past century, global surface temperatures have increased by approximately  $1.2^\circ\text{C}$  relative to the pre-industrial baseline (1850–1900), driven largely by human activity (Legg, 2021). In a warming world, extreme climate anomalies – such as heatwaves and persistent rainfall shifts – are expected to intensify (Diffenbaugh et al., 2017). These events are increasingly recognized as critical modulators of ozone variability through their impacts on photochemistry, vertical mixing, and precursor transport (Gao et al., 2023; Pu et al., 2017; Wang et al., 2022b).

Quantifying the respective roles of anthropogenic emissions and meteorological variability in driving ozone trends is therefore essential for evaluating the effectiveness of clean air policies (Li et al., 2019a; Liu et al., 2023). Previous studies have reported rapid increases in surface ozone concentrations in key Chinese regions – such as the Beijing–Tianjin–Hebei (BTH) and Yangtze River Delta (YRD) –



62 during the initial policy phase (2013 – 2017), with increases of approximately 28% and 18%, respectively  
 63 (Chen et al., 2020; Li et al., 2019a; Liu et al., 2023). In contrast, a modest decline in ozone levels was  
 64 observed during 2018 – 2020, largely attributed to emission reductions (Li et al., 2021; Liu and Wang,  
 65 2020b; Wang et al., 2024b; Wang et al., 2023a). However, since 2021, observations indicate a renewed  
 66 increase in ozone concentrations (Fig. S1). These fluctuations suggest oscillating trends over the past  
 67 decade, the drivers of which remain poorly constrained.

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

84 Here, we develop a novel machine learning-based framework – Fixed Emission Approximation  
 85 (FEA) – to quantify the respective roles of anthropogenic emissions and meteorological conditions in  
 86 shaping summertime surface ozone trends in China. Applying FEA to nationwide observational data from  
 87 2013 to 2023, we identify three distinct phases of ozone evolution corresponding to major clean air  
 88 actions and policy transitions. We further analyze short-term ozone anomalies associated with extreme  
 89 weather events, such as the 2022 heatwave and seasonal monsoon rainfall. To characterize photochemical  
 90 regimes, we integrate satellite-derived formaldehyde-to-nitrogen dioxide (HCHO/NO<sub>2</sub>, FNR) ratios from



91 Tropospheric Monitoring Instrument (TROPOMI), revealing spatiotemporal shifts in ozone formation  
92 sensitivity across China. Finally, we extend our FEA analysis to evaluate climate-driven ozone trends  
93 from 1970 to 2023, using historical meteorological reanalysis data. Together, these results provide a  
94 comprehensive picture of the anthropogenic and climatic forces shaping surface ozone dynamics in a  
95 rapidly warming and urbanizing China.

## 96 **2 Data and Methods**

### 97 **2.1 Sampling site and instruments**

98 Hourly surface ozone concentration data were obtained from the National Environmental  
99 Monitoring Center of China and can be accessed through the open website <https://air.cnemc.cn:18007/>  
100 (last accessed: May 20, 2024). Hourly meteorological data with a spatial resolution of  $0.25^\circ \times 0.25^\circ$  were  
101 sourced from the ERA5 reanalysis dataset provided by the European Centre for Medium-Range Weather  
102 Forecasts (ECMWF) and are available for download at <https://cds.climate.copernicus.eu> (last accessed:  
103 March 20, 2025). For detailed variables, refer to Table S1. The MDA8 ozone TAP dataset (Geng et al.,  
104 2021) for 2013 and 2014 can be downloaded from <http://tapdata.org> (last accessed: May 20, 2024). The  
105 Tropospheric Monitoring Instrument (TROPOMI) on the Sentinel-5P satellite provides global continuous  
106 observation data for two indicators of  $O_3$  precursor substances: nitrogen dioxide ( $NO_2$ ) and formaldehyde  
107 (HCHO) concentrations (Lamsal et al., 2014; Shen et al., 2019). The spatial resolution of TROPOMI  
108 data is 1113.2 meters (approximately  $0.009^\circ$  in China) (Ren et al., 2022).

### 109 **2.2 Machine learning-based FEA approach**

110 In this study, we propose a machine learning-based FEA approach to assess the impacts of  
111 meteorological factors and anthropogenic emissions on the year-round variations in ozone concentrations.  
112 First, we construct a regression model using the random forest (RF) algorithm to relate ozone  
113 concentrations to meteorological parameters at various atmospheric heights and to regular emission  
114 surrogate parameters (i.e., time variables). The meteorological parameters include 18 distinct variables  
115 at different altitudes, while the emission surrogate parameters include the month and the hour of the day,  
116 these temporal predictors capture the effects of day-night cycles and workday patterns on air pollutant



concentrations, reflecting the long-term trends in pollutant behavior. The aforementioned variables have been used as typical emission surrogate input features in previous studies (Grange et al., 2018; Meng et al., 2025; Shi et al., 2021; Vu et al., 2019). Our modeling strategy involves building and predicting models for individual cities and for each year from 2015 to 2023. Due to the lack of available observational data for many cities in 2013 and 2014, we did not develop models for these two years. In our approach, 80% of the dataset is used for model training, while the remaining 20% is reserved for testing. We perform ten-fold cross-validation and assess model performance using seven statistical metrics, as listed in Table S2.

Following the construction of the machine learning models for individual cities and years, we introduce the FEA approach. The key principle of FEA is the assumption that the total emissions of ozone precursors remain unchanged from the baseline year. Specifically, using the model trained on data from the baseline year ( $i$ ) as a reference for anthropogenic emissions, we establish hourly-resolution models for the summer months (June to August) of the baseline year. These models are then applied to predict ozone concentrations under the meteorological conditions of the prediction year ( $j$ ), while holding the emission levels constant at those of the baseline year ( $i$ ). The difference between the predicted values and the observed values for the baseline year ( $i$ ) represents the model residuals ( $RES_i$ ), as shown in Eq. (1). The difference in observed MDA8 ozone concentrations between two different prediction years ( $j_1, j_2$ ) is driven by the differences in meteorological conditions ( $\Delta MET_{i(j_1, j_2)}$ ) and anthropogenic emission controls ( $\Delta ANT_{i(j_1, j_2)}$ ) (Eq. 2). The term  $\Delta MET_{i(j_1, j_2)}$  represents the changes in meteorological conditions and can be calculated by the difference between the predicted values,  $Pred_{i(j_1)}$  and  $Pred_{i(j_2)}$ , for the corresponding years (Eq. 3). The prediction result  $Pred_{i(j)}$  obtained by applying the model trained with data from year  $i$  to the meteorological conditions of year  $j$  can be used to calculate the emission-driving variable  $ANT_{i(j)}$  corresponding to the model trained in year  $i$  and the meteorological conditions of year  $j$  using Eq. (4). Similarly, the value of  $\Delta ANT_{i(j_1, j_2)}$ , representing the change in anthropogenic emissions between the two years  $j_1$  and  $j_2$ , can be therefore calculated using Eq. (5). By performing these calculations, we can isolate and quantify the contributions of meteorological conditions and anthropogenic emission controls to the observed ozone trends. We used a cross-matrix research method to assess the uncertainty of FEA, with specific formulas available in Supporting Method S1.



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

$$\Delta OBS_{(j1,j2)} = \Delta MET_{i(j1,j2)} + \Delta ANT_{i(j1,j2)}, \quad (2)$$

$$\Delta MET_{i(j1,j2)} = Pred_{i(j2)} - Pred_{i(j1)}, \quad (3)$$

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

$$\begin{aligned} \Delta ANT_{i(j1,j2)} &= ANT_{i(j2)} - ANT_{i(j1)} = (OBS_{j2} - Pred_{i(j2)} - RES_i) - (OBS_{j1} - Pred_{i(j1)} - RES_i) \\ &= (OBS_{j2} - OBS_{j1}) - (Pred_{i(j2)} - Pred_{i(j1)}), \end{aligned} \quad (5)$$

Model performance was first evaluated through ten-fold cross-validation for the Beijing – Tianjin – Hebei (BTH) region, revealing high predictive skill between observed and predicted MDA8 ozone levels during 2015-2023 (Fig. S2). The index of agreement (IOA) ranged from 0.96 to 0.97, with correlation coefficients ( $R$ ) between 0.93 and 0.95. Root mean square errors (RMSE) and normalized mean bias (NMB) varied from 16.9 to 21.9  $\mu\text{g m}^{-3}$  and 8 to 25%, respectively, indicating high model accuracy. Nationally, the model yielded  $R$  values of 0.88–0.91 and IOA of 0.93–0.95, with errors remaining within acceptable ranges (Tables S3–S8). To assess uncertainty stemming from interannual model training variability, we applied a matrix-based resampling approach (Supporting Method S1). As shown in Fig. S3, the relative difference in residuals ranged from -9% to 3%, and remained within  $\pm 12\%$  for all regions – supporting the robustness of the FEA method.

### 2.3 Ozone formation regime detection with FNR

Ozone concentrations show a significant nonlinear relationship with their precursors, which can be classified into three types: the VOC-controlled zone, the  $\text{NO}_x$ -controlled zone, and the excessive/mixed zone. The ratio of HCHO to  $\text{NO}_2$  (FNR) serves as a reactive weighting of VOC/ $\text{NO}_x$  and is one of the diagnostic indicators of ozone-sensitive intervals (Sillman, 1995), this is particularly suited to the analysis of satellite data and has been widely used in related research (Jin et al., 2020; Jin and Holloway, 2015; Wang et al., 2021). Based on the framework described by Ren et al. (2022) and Jin et al. (2015), we derived a diagnostic approach that is more applicable to our data, and the present study categorizes ozone sensitivity zones for the summer of 2018-2023 according to the following criteria:

$$FNR_{avg} < 4.0 \text{ and } FNR_{avg} + FNR_{sd} < 6.0 : \text{VOC – controlled zone}$$

$$FNR_{avg} > 4.0 \text{ and } FNR_{avg} - FNR_{sd} > 2.0 : \text{NO}_x \text{ – controlled zone}$$



173 *Otherwise: excessive/mixed zones*

174 where  $FNR_{avg}$  and  $FNR_{sd}$  denote the time-mean and standard deviation of the FNR for the target  
 175 time period.

#### 176 **2.4 FEA-based assessment of climate change impacts on ozone**

177 To further evaluate the long-term impact of climate change on ozone concentrations over China  
 178 from 1970 to 2023, we extended the framework of our proposed FEA method. The core idea of this  
 179 analysis is to isolate the influence of long-term meteorological variations on ozone, assuming fixed  
 180 anthropogenic emissions. Given the availability of relatively complete and continuous hourly ozone  
 181 observations and meteorological data across China from 2015 to 2023, we selected this period as the  
 182 basis for constructing emission baselines.

183 Following the modeling protocol described in the section Machine learning-based FEA, we trained  
 184 nine separate random forest models – each using a different year from 2015 to 2023 as an emissions  
 185 reference. Inputs included hourly ozone observations, key meteorological predictors, and time-related  
 186 variables (hour of day and month of year). These trained models were then applied to historical reanalysis  
 187 meteorology from 1970 to 2023 to simulate ozone trends under constant emissions. This yielded nine  
 188 independent ozone trajectories, each reflecting the influence of long-term meteorological variability  
 189 under a different fixed-emissions assumption.

190 While the choice of emission baseline may affect the absolute magnitude of simulated ozone, it does  
 191 not alter the primary objective: assessing the sensitivity of surface ozone to meteorological drivers over  
 192 multidecadal timescales (Lecœur et al., 2014; Leung et al., 2018; Wang et al., 2022c). This approach  
 193 captures the climate-induced ozone signal while adopting the commonly used assumption that emissions  
 194 are not themselves influenced by climate change – a simplification consistent with prior attribution  
 195 studies (Dang and Liao, 2019; Leung et al., 2018; Shen et al., 2017; Wang et al., 2022c). For comparison,  
 196 we also estimated the impact of anthropogenic emission changes on ozone concentrations during the  
 197 observational window of 2015–2023, using the same FEA methodology and the complete hourly dataset  
 198 for model training. This dual-track analysis enables a clear distinction between the contributions of  
 199 climate variability and emission mitigation to observed ozone trends.





## 200     **3 Results and Discussion**

### 201     **3.1 Spatiotemporal Evolution of Summertime Ozone (2013 – 2023)**

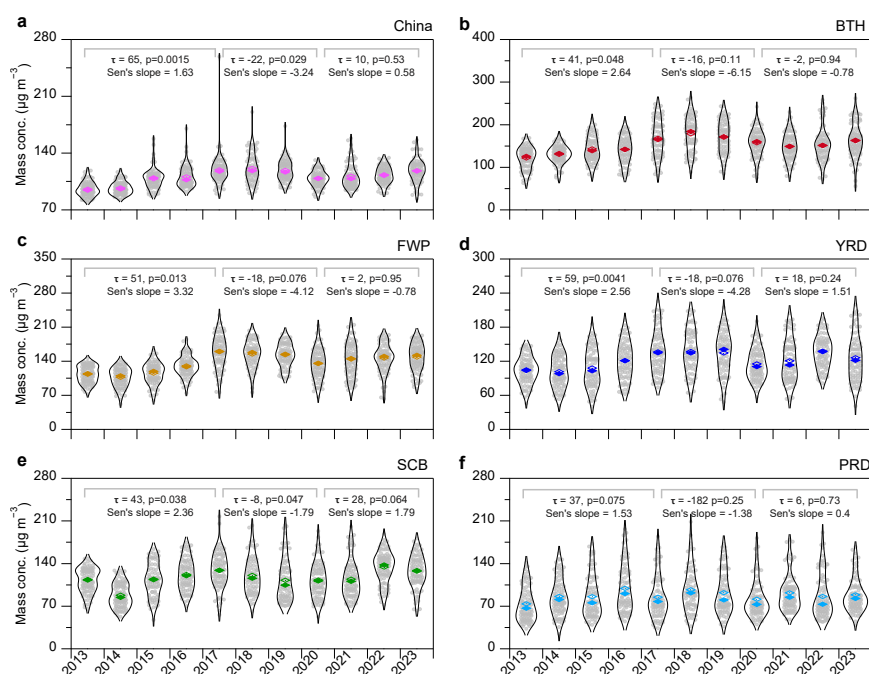
202         Figure 1 presents the interannual variations in maximum daily 8-hour average (MDA8) ozone  
 203         concentrations during summertime (June–August) across China, with a focus on five key urban  
 204         agglomerations: Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD), Fenwei Plain (FWP),  
 205         Sichuan Basin (SCB), and Pearl River Delta (PRD). From 2013 to 2023, summertime ozone levels  
 206         displayed distinct temporal patterns across regions, reflecting the impact of successive national emission  
 207         control phases. During the first phase (2013–2017), nationwide MDA8 ozone increased significantly ( $p$   
 208          $< 0.01$ ), rising from 95.5 to 118.0  $\mu\text{g m}^{-3}$ . This growth was especially pronounced in the BTH and FWP  
 209         regions, where concentrations increased by 38% and 41%, respectively. In contrast, ozone increases were  
 210         more modest in the YRD (11%), SCB (15%), and PRD (16%) regions, respectively. These results were  
 211         consistent with the previous studies (Li et al., 2021; Liu and Wang, 2020a, b; Wang et al., 2023a).

212         In the second phase (2017–2020), corresponding to the implementation of more stringent emission  
 213         controls on  $\text{NO}_x$  and VOCs emissions (Geng et al., 2024; Liu et al., 2023), a moderate national decrease  
 214         in MDA8 ozone was observed, with concentrations declining to 109.0  $\mu\text{g m}^{-3}$ . The regional declines  
 215         during this period were most notable in FWP (–16%) and YRD (–15%), while BTH (–6%), SCB (–11%),  
 216         and PRD (–4%) also showed reductions compared to their concentration peaks observed in 2017.  
 217         However, this downward trend did not persist. In the third phase (2020–2023), the MDA8 ozone  
 218         rebounded, reaching 118.4  $\mu\text{g m}^{-3}$  in 2023 – comparable to its 2017 peak – with a particularly sharp  
 219         increase during the summer of 2022. From 2021 to 2023, MDA8 ozone concentrations rose by 2.8  $\mu\text{g m}^{-3}$   
 220         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.

221         Figure S1 further illustrates the spatiotemporal evolution of summertime MDA8 ozone across 354  
 222         cities in China from 2013 to 2023. On average, 68% of cities exceeded the World Health Organization  
 223         (WHO) air quality guideline of 100.0  $\mu\text{g m}^{-3}$  for the MDA8 ozone. Elevated ozone levels were primarily  
 224         observed in densely populated and economically developed eastern regions, such as North China Plain.  
 225         Across the five major city clusters, the average ozone levels ranged from 89.4 to 152.8  $\mu\text{g m}^{-3}$  –  
 226         substantially exceeding the 43.0  $\mu\text{g m}^{-3}$  threshold associated with ecosystem productivity loss (Gong et  
 227         al., 2021), implying significant threats to both human and ecological health. Spatially, ozone hotspot



regions expanded between 2013 and 2017 (Fig. S1 a-e), followed by contraction during 2018-2020 (Fig. S1 f-i), reflecting initial policy effectiveness. However, this progress stalled from 2021. A sharp reversal was observed in 2022, with widespread increases in MDA8 ozone (Fig. S1 k), suggesting that the influence of emerging meteorological extremes or evolving ozone photochemical regimes may be counteracting the gains from emission reductions.



**Figure 1. Interannual trends of summertime MDA8 ozone across China (2013–2023).** Panel (a) illustrate the seasonal variations of MDA8 ozone during the summer months (June, July, and August) across 354 cities nationwide. Panels (b-f) shows the average trend across five key regions in China: Beijing-Tianjin-Hebei (BTH), Fenwei Plain (FWP), Yangtze River Delta (YRD), Sichuan Basin (SCB), and Pearl River Delta (PRD). The summer months are defined according to meteorological seasonality, encompassing June, July, and August. In the violin plots, hollow diamond markers denote the mean, while solid diamond markers represent the median. The Mann-Kendall test and Sen's slope estimator were employed to assess the statistical significance and rate of change in the monthly average MDA8 ozone concentrations.

### 3.2 Anthropogenic drivers of ozone trends

To isolate the influence of anthropogenic emissions on summertime ozone variability, we implemented a machine learning-based FEA framework (Sect. 2.2). This framework employs random forest (RF) models to disentangle the respective contributions of emission changes and meteorological



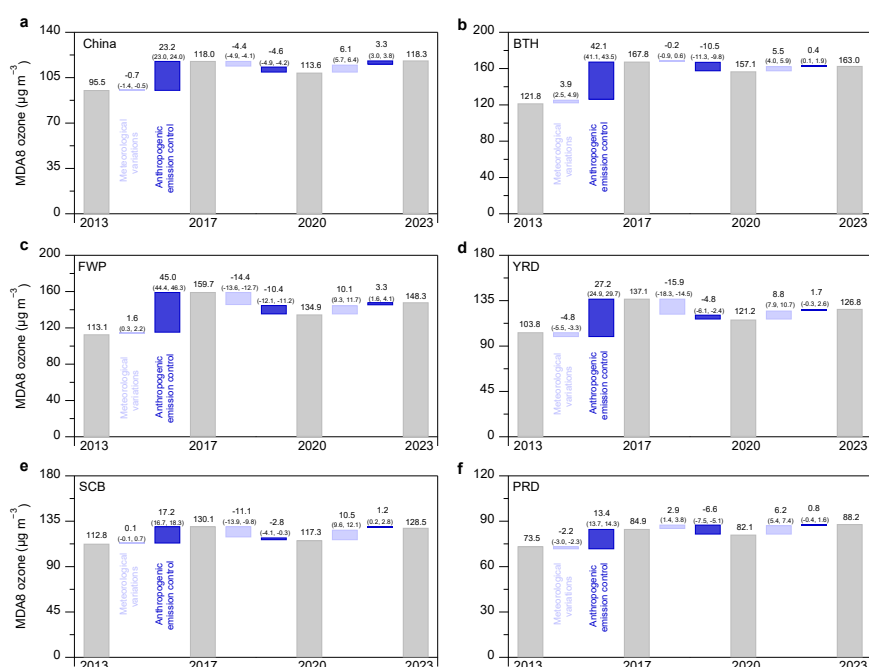
variability to observed ozone trends. As illustrated in Fig. 2, anthropogenic emissions were the dominant driver of ozone increases during 2013 – 2017, contributing an average rise of  $23.2 \pm 1.1 \mu\text{g m}^{-3}$  across 354 cities over China. The strongest regional increases occurred in the FWP and BTH, with contributions of  $45.0 \pm 2.0 \mu\text{g m}^{-3}$  and  $42.1 \pm 2.0 \mu\text{g m}^{-3}$ , respectively. In contrast, the PRD exhibited a smaller increase ( $13.4 \pm 1.6 \mu\text{g m}^{-3}$ ). These findings indicate that the emission control strategies during China's first phase of air quality control efforts, which primarily focused on reducing  $\text{PM}_{2.5}$  and haze, inadvertently contributed to worsening ozone pollution by altering the atmospheric chemistry and precursor balance (Zhang et al., 2019; Zheng et al., 2018). This finding is consistent with previous model-based assessments using chemical transport models (Li et al., 2021; Wu et al., 2022), and supports the reliability of the FEA framework in attributing observed ozone changes to underlying drivers.

In 2018, China launched its second-phase Clean Air Action Plan, which aimed to the coordinated control of both  $\text{PM}_{2.5}$  and ozone by reducing  $\text{NO}_x$  and VOCs emissions (Zhang et al., 2019; Zheng et al., 2018). During this period (2017–2020), summertime ozone concentrations decreased substantially in northern China. As shown in Fig. 2, the MDA8 ozone declined by  $10.5 \pm 2.0 \mu\text{g m}^{-3}$  in BTH and  $10.4 \pm 3.0 \mu\text{g m}^{-3}$  in FWP, with smaller but consistent declines in YRD ( $-4.8 \pm 3.8 \mu\text{g m}^{-3}$ ), SCB ( $-2.8 \pm 2.4 \mu\text{g m}^{-3}$ ), and PRD ( $-6.6 \pm 1.4 \mu\text{g m}^{-3}$ ) during 2017 – 2020. These changes underscore the effectiveness of targeted precursor controls and align well with prior studies (Liu et al., 2023; Wang et al., 2023a).

This period also overlapped with the COVID-19 pandemic, which occurred from January to April 2020, introduced an unprecedented, large-scale perturbation to human activity. The nationwide lockdown led to dramatic declines in industrial production, energy consumption, and transportation (Shi and Brasseur, 2020; Zheng et al., 2021). This provided a natural experiment to evaluate the short-term ozone response to abrupt anthropogenic emission reductions. As shown in Fig. S4, from 2017 to 2020, the MDA8 ozone annual mean levels showed a slight national decline, but the pandemic led to an increase in BTH, FWP, YRD, and SCB by  $+1.7$  to  $+2.3 \mu\text{g m}^{-3}$ , while PRD experienced a decline. Further analysis (Fig. S5) indicates that  $\sim 79\%$  of cities saw increases in ozone during this period, with a national average rise of  $2.1 \pm 1.3 \mu\text{g m}^{-3}$ . These increases are consistent with suppressed NO titration and enhanced photochemical ozone production under cleaner atmospheric conditions (Shi et al., 2021; Wang et al., 2022a). In the post-pandemic period (2020–2023), the influence of anthropogenic emissions on



summertime ozone trends became more subdued Emission-driven changes showed relatively small and mixed contributions across all regions, ranging from  $-1.2$  to  $+2.6 \mu\text{g m}^{-3}$  in BTH,  $-1.6$  to  $+4.0 \mu\text{g m}^{-3}$  in FWP,  $-4.7$  to  $+7.4 \mu\text{g m}^{-3}$  in YRD,  $-3.6$  to  $+3.0 \mu\text{g m}^{-3}$  in SCB, and  $-3.8$  to  $+7.7 \mu\text{g m}^{-3}$  in PRD (Fig. S6). These limited impacts suggest that the benefits of prior emission reduction efforts may have plateaued, and that other drivers – particularly meteorological extremes – are becoming increasingly prominent in shaping ozone variability.



**Figure 2. Anthropogenic and meteorological drivers of ozone trends from 2013 to 2023.** Changes in summertime MDA8 ozone concentrations were decomposed into contributions from anthropogenic emissions and meteorological variability using the FEA framework. Results reflect ensemble estimates based on multiple baseline years (2015–2023) for emissions. Boxplots indicate the interquartile range, with values in parentheses denoting the 25th and 75th percentiles across all baseline scenarios.

### 3.3 Ozone formation sensitivity and regime shifts

To diagnose the chemical sensitivity of ozone formation, we analyzed the spatial distributions of tropospheric  $\text{NO}_2$  and  $\text{HCHO}$  columns retrieved by TROPOMI during summer months from 2018 to 2023 (Fig. S7–S8).  $\text{NO}_2$  concentrations displayed strong spatial gradients, with eastern China exhibiting levels five times higher than the west – reflecting dense population centers and elevated anthropogenic



294 NO<sub>x</sub> emissions. While NO<sub>2</sub> levels steadily declined over time, the summertime average NO<sub>2</sub> column  
 295 concentration in the North China Plain decreased from  $4.13 \times 10^{15}$  molecules cm<sup>-2</sup> in 2018 to  $3.85 \times 10^{15}$   
 296 molecules cm<sup>-2</sup> in 2023, HCHO concentrations remained relatively stable during 2018–2021. However,  
 297 a sharp increase in HCHO was observed in the Yangtze River Delta during the record-breaking heatwave  
 298 of 2022, likely due to elevated biogenic and anthropogenic VOC emissions under extreme temperatures  
 299 (Qin et al., 2025; Tao et al., 2024). By 2023, HCHO levels returned to near-baseline, consistent with  
 300 cooler summer conditions.

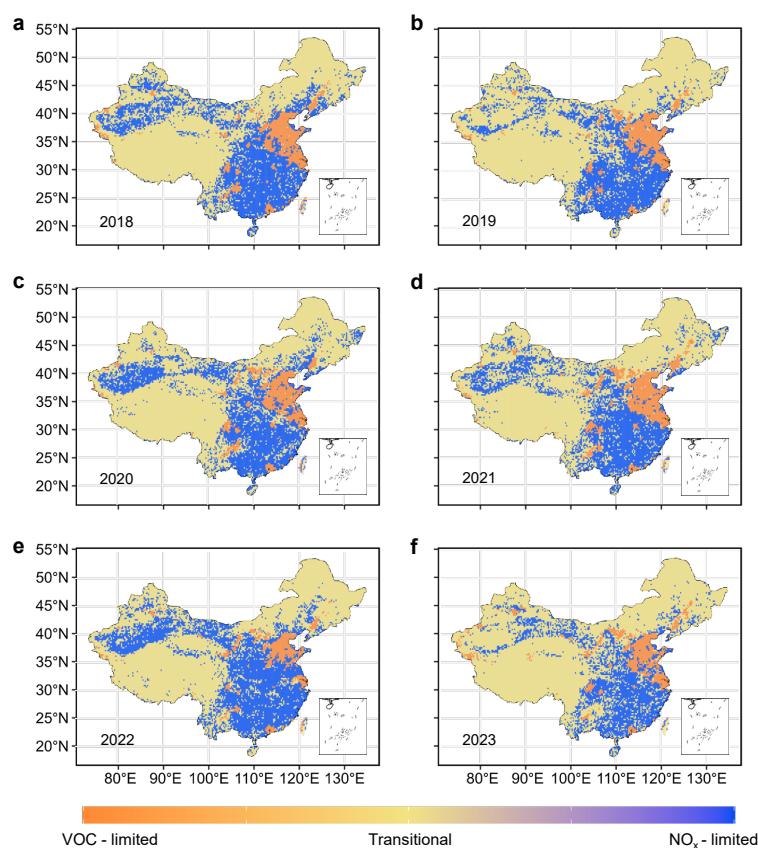
301 To further characterize the photochemical regimes, we derived the threshold of the HCHO/NO<sub>2</sub> ratio  
 302 (FNR), a widely used proxy for ozone formation sensitivity (Jin and Holloway, 2015; Li et al., 2024; Ren  
 303 et al., 2022; Wang et al., 2021). As shown in Fig. 3, extensive VOC-limited and transition zones were  
 304 observed in major megacity clusters. most urbanized regions of China remained within the VOC-limited  
 305 throughout the study period, with the notable exception of the PRD, which was predominantly NO<sub>x</sub>-  
 306 limited or transitional regimes. This is consistent with previous studies, where VOC-limited regimes  
 307 primarily appeared in economically developed and densely populated urban areas, with transition zones  
 308 surrounding VOC-limited areas in large cities and suburbs (Li et al., 2024; Shen et al., 2021).

309 From 2018 to 2020, regime boundaries exhibited only modest changes. During this period, the  
 310 VOC-limited areas in the study region gradually decreased, while transition zones correspondingly  
 311 increased. Additionally, some areas initially classified as transitional regimes shifted to NO<sub>x</sub>-limited  
 312 regimes. The expansion of mixed and NO<sub>x</sub>-limited regimes was closely associated with significant NO<sub>x</sub>  
 313 emission reductions (Wang et al., 2023a). In 2021, the VOC-limited area expanded slightly across eastern  
 314 China. A more dramatic shift occurred in 2022, as extreme heat and elevated VOC levels drove  
 315 widespread transitions from VOC-limited to transitional or NO<sub>x</sub>-limited regimes, especially across the  
 316 YRD and surrounding regions.

317 Monthly regime evolution from 2020 to 2023 (Fig. S9) confirms that the most extensive regime  
 318 shifts occurred in August 2022 (Fig. S9i), coinciding with peak temperatures and FNR anomalies.  
 319 Notably, VOC-limited areas tended to be smaller in July and August compared to June, likely due to  
 320 increased VOC reactivity under higher temperatures (Fig. S9). However, major cities generally remained  
 321 VOC-limited, while adjacent suburban areas shifted dynamically between transitional and VOC-limited  
 322 regimes. In contrast, outer suburbs and rural regions were more frequently controlled by NO<sub>x</sub> (Shen et



al., 2021; Wang et al., 2021). Although VOC-limited regimes partially recovered in 2023, their spatial extent remained smaller than in 2021, likely due to ongoing  $\text{NO}_x$  emission reductions outpacing changes in VOCs emissions, contributing to a structural shift in ozone formation chemistry. These findings highlight the influence of climate-induced VOCs responses and precursor imbalance in driving ozone formation regime shifts and complicating ozone mitigation efforts. While this influence has already become prominent in the current phase, it is expected to intensify with the increasing frequency of extreme weather events in the future.



**Figure 3. Ozone formation sensitivity regimes.** The results of FNR analysis from June to August (2018-2023) are presented, showing the spatiotemporal variation of ozone sensitivity in different regions. The colors in the map represent the geographical distribution of VOC-limited,  $\text{NO}_x$ -limited, and transitional ozone sensitivity zones. The city locations within the five key regions of China are shown in Fig. S10.

### 3.4 Impact of meteorological variations on ozone



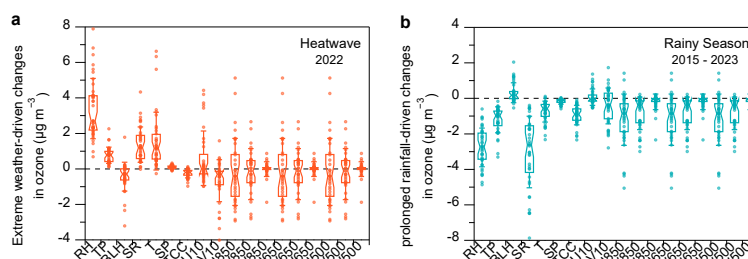
337 Meteorological conditions directly and indirectly modulate surface ozone concentrations by  
 338 influencing photochemical reactions, vertical mixing, and dispersion processes (Li et al., 2019b; Li et al.,  
 339 2020). These effects exhibit strong regional and temporal heterogeneity across China. As shown in Fig.  
 340 2, during Phase I (2013–2017), meteorological contributions to summertime MDA8 ozone remained  
 341 modest, ranging from  $-4.8$  to  $+3.9 \mu\text{g m}^{-3}$ . In Phase II (2017–2020), notable ozone reductions attributable  
 342 to meteorology were observed  $-14.4 \pm 3 \mu\text{g m}^{-3}$ ,  $-15.9 \pm 3.8 \mu\text{g m}^{-3}$ , and  $-11.1 \pm 2.4 \mu\text{g m}^{-3}$  in FWP,  
 343 YRD, and SCB, respectively. These reductions accounted for  $58 \pm 12\%$ ,  $77 \pm 18\%$ , and  $80 \pm 17\%$  of the  
 344 total summertime MDA8 ozone reduction during this phase. however, these impacts remained smaller  
 345 than those from emission controls in BTH and PRD. In contrast, during 2020–2023, ozone trends became  
 346 increasingly influenced by meteorological anomalies, particularly in 2022. That summer, extreme  
 347 heatwaves (Mallapaty, 2022; Wang et al., 2023b) led to sharp increases in MDA8 ozone, contributing  
 348  $20.8 \pm 3.6 \mu\text{g m}^{-3}$  in YRD and  $22.1 \pm 3.2 \mu\text{g m}^{-3}$  in SCB. In 2023, however, abundant summer rainfall  
 349 suppressed ozone formation, with MDA8 ozone decreasing by  $-17.8 \pm 2.3 \mu\text{g m}^{-3}$  in YRD and  $-9.7 \pm 3.3$   
 350  $\mu\text{g m}^{-3}$  in SCB. These declines correspond to year-on-year increases in rainfall of 102% and 35% in the  
 351 two regions, respectively (Fig. S11).

352 To further elucidate the dominant meteorological drivers of ozone variability, we examined Gini  
 353 importance (Nembrini et al., 2018; Wright and Ziegler, 2017) scores derived from the RF model across  
 354 18 predictor variables (Fig. S12). Temperature ( $T$ ) and relative humidity (RH) emerged as the most  
 355 influential variables in the BTH, FWP, and SCB regions, while in the YRD, shortwave solar radiation  
 356 (SR), RH, and rainfall were dominant. These results suggest that ozone variability is governed by  
 357 complex meteorological interactions that vary regionally. For instance, rainfall is typically associated  
 358 with lower solar irradiance and increased cloud cover, both of which are unfavorable for photochemical  
 359 ozone production (Jacob and Winner, 2009; Shan et al., 2008). Moreover, the high importance of  $T$  and  
 360 SR in these regions indicates that surface ozone is highly sensitive to thermal conditions and  
 361 photochemical intensity (Yang et al., 2025). Elevated temperatures accelerate ozone precursor emissions  
 362 and reaction rates, while stronger solar radiation enhances photolysis and ozone formation potential (Qin  
 363 et al., 2025; Tao et al., 2024). In the PRD, ozone variability was more strongly influenced by temperature  
 364 and transport-related indices (such as meridional winds at different layers, etc.). This likely reflects the  
 365 region's subtropical coastal climate, where frequent summer typhoon incursions from the Northwest



Pacific modulate large-scale atmospheric transport (Chen et al., 2024; Wang et al., 2024a; Wang et al., 2022b). These events may introduce strong horizontal advection and vertical mixing, thereby altering the distribution and buildup of ozone precursors, and contributing significantly to the observed ozone variability. As shown in Fig. S13, the correlations between ozone and key meteorological variables were notably enhanced during heatwave (HW) periods. Specifically, ozone positively correlated with both  $T$  and SR, and negatively (or weakly) correlated with RH. During prolonged rainfall (PR) events, cities in the Yangtze-Huaihe region showed the strongest RH–ozone anti-correlation ( $R < -0.7$ ), likely driven by the enhanced wet scavenging and reduced photochemistry (Fig. S14 a – c).

To quantify the individual contributions of meteorological variables, we applied SHAP (SHapley Additive exPlanations) analysis to HW and PR events in the Yangtze-Huaihe region from 2015 to 2023 (Supporting Methods S3). As shown in Fig. S15 and Fig. S14 d, HW events were associated with strong positive SHAP values in southeastern coastal cities, the YRD, and SCB – primarily driven by elevated SR and  $T$ . Indeed, mean SR during HW periods was significantly higher than during non-HW periods (Fig. S16), amplifying photochemical ozone production potential. In contrast, PR events consistently yielded negative SHAP contributions across all cities, mainly due to reduced sunlight and suppressed precursor buildup. A multi-year comparison (Fig. 4) highlights the opposing effects of key meteorological variables – including RH,  $T$ , boundary layer height, total precipitation, and surface pressure – on MDA8 ozone. SR, RH, and  $T$  emerged as the most influential parameters, while total cloud cover and meteorological transport playing secondary roles during HW episodes. The intensity of HW and PR events modulated the magnitude of these effects. For instance, high-rainfall PR events in 2016 and 2020 yielded large negative SHAP contributions ( $-29.7$  and  $-16.9 \mu\text{g m}^{-3}$ ), mainly via RH-driven suppression. Conversely, reduced rainfall in 2023 weakened the RH effect, though advection and vertical mixing still contributed to ozone suppression (Fig. S17).



**Figure 4. Meteorological influences on predicted ozone concentrations under heatwave and rainy weather conditions.** (a) Differences in SHAP values ( $\Delta\text{SHAP}$ ) between heatwave and non-heatwave periods in the Yangtze-





392 Huaihe region during summer 2022. (b) Differences in SHAP values ( $\Delta$ SHAP) between prolonged rainfall periods  
 393 and non-prolonged rainfall periods in the same region from 2015 to 2023. Box plots show the distribution of  $\Delta$ SHAP  
 394 across cities; the center line indicates the median, boxes denote the interquartile range (25th-75th percentiles), and  
 395 whisker line extends to one standard deviation.

396

### 397 **3.5 Reshaping ozone trends in a warming climate**

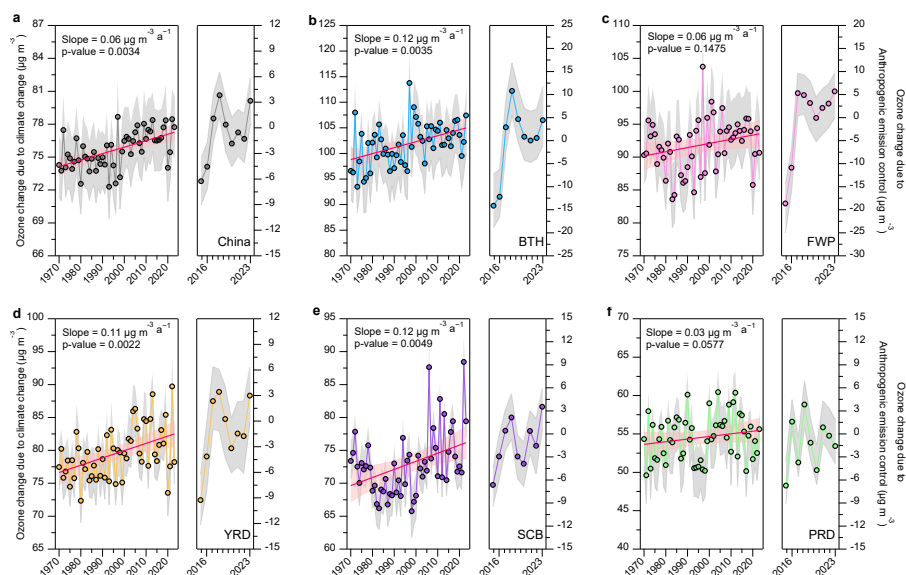
398 To assess the long-term influence of climate change on surface ozone concentrations, we applied  
 399 the FEA framework to simulate summertime ozone trends over the period 1970 – 2023. In this analysis,  
 400 anthropogenic emissions were held constant at their 2015 – 2023 summertime levels, while interannual  
 401 variations in meteorological variables were introduced using historical reanalysis data. This design  
 402 isolates the climate-driven component of ozone trends while assuming that emission trajectories are  
 403 independent of climate change – a simplification aligned with prior attribution frameworks (Wang et al.,  
 404 2022c). The impact of anthropogenic emission controls was estimated by comparing observed ozone  
 405 concentrations with FEA-predicted values during 2015 – 2023, thereby quantifying the residual effect of  
 406 emissions under fixed meteorology.

407 As shown in Fig. 5, under the 2015-2023 emission levels, climate change has exerted a statistically  
 408 significant ( $p < 0.05$ ) positive influence on urban summertime ozone concentrations across China,  
 409 resulting in a nationwide increase of approximately  $0.06 \mu\text{g m}^{-3} \text{a}^{-1}$  since 1970. All five major urban  
 410 regions displayed upward trends, with the most pronounced increase observed in the BTH and SCB at  
 411  $0.12 \mu\text{g m}^{-3} \text{a}^{-1}$ . Spatial correlations between climate-driven ozone increases and temperature changes  
 412 (Fig. S18) further confirm that warming is the dominant contributor to long-term ozone enhancement. In  
 413 particular, the correlation coefficients between ozone trends and temperature anomalies reached 0.90  
 414 (BTH), 0.89 (FWP), 0.72 (YRD), and 0.93 (SCB), indicating a strong temperature dependence of  
 415 climate-induced ozone formation in these regions. The PRD showed a weaker correlation, likely due to  
 416 its unique subtropical maritime climate and higher humidity and cloud cover, which tend to suppress  
 417 photochemical ozone production (Yang et al., 2019).

418 These findings are consistent with previous projections that forecast an increase in high-ozone  
 419 events under future climate scenarios spanning 2020–2100 (Li et al., 2023). The historical record already  
 420 reflects this risk: despite significant increases in anthropogenic emissions driving ozone growth prior to  
 421 2018, national air quality improvement measures began to yield reductions thereafter. However, since



2020, a rebound in ozone concentrations has emerged in several regions, suggesting that the climatic penalty for ozone is beginning to offset the benefits of emission control. The intense heatwave of the 2022 summer significantly enhanced ozone formation and altered ozone sensitivity – shifting from a VOC-limited region to a transition zone or NO<sub>x</sub>-limited region. Meanwhile, the reduction in anthropogenic emissions of ozone precursor substances indirectly led to changes in ozone sensitivity, thereby making anthropogenic emission reductions more effective in ozone control. However, overall, the direct effects of climate change (i.e., increased ozone formation) far outweigh the indirect effects of anthropogenic emission controls, indicating that the punitive effects of climate change on ozone will become increasingly significant in the future. Taken together, these results underscore the dual challenges of air quality management in a warming climate. Anthropogenic emission reductions remain critical, but they may no longer suffice in isolation. As the warming-driven enhancement of ozone formation becomes more prominent, China and other rapidly urbanizing regions will require adaptive and climate-resilient air quality strategies – including dynamic precursor control, land-use planning, and extreme weather early warning systems –to sustainably mitigate ozone pollution in the decades to come.



**Figure 5. Impact of climate change and emission controls on ozone trends.** Left panels show ozone trends attributable to long-term climate change from 1970 to 2023, simulated under fixed emission scenarios using the FEA framework. Right panels depict ozone trends from 2015 to 2023, reflecting the impact of anthropogenic emission controls. Each trajectory represents results based on a distinct emissions baseline year. Shaded grey areas indicate the interquartile range (25th-75th percentiles), solid red lines denote trend estimates, and light red shading marks the



442 5th-95th percentile confidence intervals. Statistical significance and trend slopes were assessed using the Mann-  
 443 Kendall test.

444

#### 445 **4 Conclusions and implications**

446 China is confronted with the dual challenges of climate change and ozone pollution. Over the  
 447 past decade, summertime ozone concentrations across the country have exhibited complex  
 448 spatiotemporal patterns, reflecting the evolving interplay between anthropogenic emissions,  
 449 meteorological variability, and large-scale climate dynamics. In this study, we developed and  
 450 applied a machine learning-based FEA framework to disentangle and quantify the respective roles  
 451 of anthropogenic emissions and meteorological drivers in shaping ozone trends during 2013–2023.  
 452 With a national-level prediction uncertainty of approximately 6%, the FEA method provides a  
 453 computationally efficient and scalable tool for diagnosing atmospheric variability across large  
 454 spatial and temporal domains.

455 Our analysis revealed that increased anthropogenic precursor emissions were the dominant  
 456 driver of the sharp rise in summertime MDA8 ozone concentrations during the first phase (2013–  
 457 2017), contributing an average increase of  $23.2 \pm 1.1 \mu\text{g m}^{-3}$ . In contrast, during the second phase  
 458 (2018–2020), enhanced air quality regulations – particularly the synergistic control of  $\text{NO}_x$  and  
 459 VOCs – led to measurable reductions in MDA8 ozone, with national-average declines of  $4.6 \pm 1.5$   
 460  $\mu\text{g m}^{-3}$ . These improvements were especially evident in regions such as BTH and FWP, where ozone  
 461 formation is highly sensitive to VOC levels. However, during the most recent period (2021–2023),  
 462 the impact of emission reductions diminished considerably, with regional ozone levels either  
 463 plateauing or rebounding. This stagnation underscores the urgent need for more targeted, region-  
 464 specific emission control strategies that address the shifting photochemical sensitivity of ozone  
 465 formation regimes.

466 Applying the SHAP method, we further quantified the impacts of extreme meteorological  
 467 events on ozone levels. Our results show that record-breaking heatwaves in 2022 contributed to  
 468 widespread ozone enhancements of up to  $5.8 \mu\text{g m}^{-3}$ , while prolonged rainfall events – particularly  
 469 during the East Asian plum rain seasons – suppressed ozone production by as much as  $-15.2 \mu\text{g m}^{-3}$ .



470 These findings highlight the increasingly dominant role of short-term meteorological extremes in  
471 modulating ozone air quality under a warming climate. In parallel, satellite-based FNR analysis  
472 diagnostics revealed that most urban clusters in China remained in VOC-limited or transitional  
473 regimes, except the PRD, which was largely NO<sub>x</sub>-limited. The 2022 heatwave triggered regime  
474 shifts in regions such as the YRD, where rising VOCs emissions and elevated temperatures shifted  
475 the photochemical regime toward NO<sub>x</sub>-limited. These results emphasize the importance of dynamic,  
476 region-specific assessments of ozone formation sensitivity in the formulation of effective mitigation  
477 strategies.

478 To assess the climate penalty on ozone, we extended the FEA framework to simulate long-term  
479 trends from 1970 to 2023, by fixing emissions and allowing meteorological variables to evolve with  
480 observed climate trends. Our findings show that climate change has contributed a significant upward  
481 trend in urban summertime ozone, averaging 0.06 μg m<sup>-3</sup> a<sup>-1</sup>, with particularly strong increases in  
482 the BTH and SCB. Correlations between ozone and surface temperature were consistently high  
483 ( $r = 0.72\text{--}0.93$ ) in BTH, FWP, YRD, and SCB, suggesting that warming has increasingly offset gains  
484 from emission controls in recent years.

485 While the FEA framework provides a powerful diagnostic tool, some limitations remain. For  
486 example, the historical simulations did not account for climate-driven changes in land use,  
487 topography, or population density, which may introduce biases in long-term attribution (Zhu et al.,  
488 2025). Future work could incorporate dynamic ancillary datasets and emissions scenarios to further  
489 improve model performance. Overall, this study underscores the escalating influence of climate  
490 extremes on ozone variability and the emerging limits of conventional emission control approaches.  
491 In the face of continued warming, machine learning-based attribution frameworks such as FEA offer  
492 a promising pathway for integrating meteorology, chemistry, and policy analysis. To achieve  
493 sustained improvements in ozone air quality, future strategies must consider the compound effects  
494 of anthropogenic emissions, short-term weather events, and long-term climate change, and adopt  
495 adaptive, region-specific, and climate-resilient air quality management frameworks.

496



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

498

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

501

502 *Author contributions.*

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

506

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

508

509 *Acknowledgments.*

510 This study was supported by the National Natural Science Foundation of China (grant no. 42207124)  
 511 and Natural Science Foundation of Jiangsu Province (grant no. BK20210663).

512

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