

Meteorological influence on surface ozone trends in China: Assessing uncertainties caused by multi-dataset and multi-method

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Abstract. China has witnessed notable increases in surface ozone (O₃) concentrations since 2013, with meteorology identified as a critical driver. However, meteorological contributions vary with different meteorological datasets and analytical methods, and their uncertainties remain unassessed. This study leveraged decadal observational maximum daily 8-hour average O₃ records (2013–2022) across China, revealing intensified nationwide O₃ pollution with increasing O₃ trends of 0.79–1.31 ppb yr⁻¹ during four seasons. We gave special focus on uncertainties of meteorology-driven O₃ trends by using diverse meteorological datasets (ERA5, MERRA2, FNL) and diverse analytical methods (Multiple Linear Regression, Random Forest, GEOS-Chem model). A useful statistic (coefficient of variation, CV) was adopted as an uncertainty quantification metric. For multi-dataset analysis, models driven by different meteorological datasets exhibited the maximum meteorology-driven O₃ trend (+0.55 ppb yr⁻¹, multi-dataset mean) with the highest consistency (CV=0.25) in spring. The FNL-driven model always obtained larger trends compared to ERA5 and MERRA2, which could be attributed to inability to accurately evaluate planetary boundary layer height in FNL dataset. For multi-method analysis, three methods demonstrated optimal consistency in winter (CV=0.40) and the worst consistency in summer (CV=2.00). The meteorology-driven O₃ trends obtained from GEOS-Chem model were almost smaller than those obtained by other two methods, partly resulting from higher simulated O₃ values before 2018. Overall, all analyses driven by diverse meteorological datasets and analytical methods drew a robust conclusion that meteorological conditions almost boosted O₃ increases during all seasons; the uncertainties caused by different analytical methods were larger than those caused by diverse meteorological datasets.

Keywords: Meteorological impact, O₃ trend, Uncertainty

30 1 Introduction

31 Since 2013, the Chinese government has implemented a series of policies to mitigate air pollution resulting from the ~~repaid~~
32 rapid industrial and urban expansion, such as the “Air Pollution Prevention and Control Action Plan” (Wang, 2021). Several
33 criteria air pollutants exhibited decreases due to the emission control efforts, but not ozone (O₃) (Qi et al., 2023; Shen et al.,
34 2020). In China, O₃ concentrations were increased by 50–124 µg m⁻³ from 2015 to 2022 (Yao et al., 2024). The formation of
35 surface O₃ depends nonlinearly on its precursors and is strongly influenced by meteorological conditions and anthropogenic
36 emissions (Wang et al., 2017). The impact of emission-related factors on O₃ increase in China over the past decade has been
37 extensively debated, including the ineffective control of volatile organic compounds (VOCs) emissions, the heightened O₃
38 photochemical production due to the rapid decrease in PM_{2.5}, and the reduced nitric oxide (NO) titration effect (Li et al., 2019,
39 2022; Lin et al., 2021a; Liu and Wang, 2020b; Ren et al., 2022).

40 Meteorological conditions also play a crucial role in shaping surface O₃ trends (Liu et al., 2023; Lu et al., 2019b), resulting in
41 increased O₃ concentrations during warm seasons over most of the United States, the European Union, and China from 2014
42 to 2019 (Lyu et al., 2023). In China, the meteorological impacts on O₃ levels may be comparable to the anthropogenic
43 contributions (Li et al., 2020; Liu and Wang, 2020a). From 2013 to 2018, meteorology could account for 43% of the daily
44 variability in summer surface O₃ concentrations in eastern China (Han et al., 2020). Adverse meteorological conditions were
45 identified as the cause of the worsening O₃ trends during 2015–2020 in Beijing-Tianjin-Hebei (BTH), Yangtze River Delta
46 (YRD), and Pearl River Delta (PRD) regions (Hu et al., 2024b). In YRD, Dang et al. (2021) found that meteorological factors
47 contributed 84% of the O₃ increase during the summers of 2012–2017. In PRD, meteorological conditions contributed 83% of
48 the increasing O₃ trends during the summers of 2015–2019 (Mousavinezhad et al., 2021). After 2019, meteorological
49 conditions tended to improve O₃ air quality (Liu et al., 2023; Wang et al., 2023). Compared to 2019, the wetter and cooler
50 meteorological conditions in 2020 reduced O₃ concentrations by 2.9 ppb in eastern China (Yin et al., 2021). However, during
51 2022’s summer, a notable rebound in O₃ levels occurred with O₃ concentrations rising by 12–15 ppb in China compared to
52 2021, which was attributable to the extreme heatwave events (Qiao et al., 2024). With climate change, the frequency of extreme
53 O₃ pollution events is expected to increase (Gao et al., 2023; Ji et al., 2024). Given the shifted meteorological effects on O₃
54 and climate change, it is imperative to conduct O₃-Meteorology researches focusing on longer time frames to gain deeper
55 insights into the long-term changes in O₃ concentrations (Wang et al., 2024a).

56 Studies conducted over the past six years to determine the meteorological influence on the surface O₃ trend have been
57 systematically reviewed, as documented in [Table S1](#). The meteorological influence on surface O₃ concentrations is commonly
58 assessed by using the traditional statistical model (TSM), machine learning model (MLM), and chemical transport model
59 (CTM), driven by reanalysis meteorological products such as the fifth-generation European Centre for Medium-Range
60 Weather Forecasts atmospheric reanalysis of the global climate (ERA5), the Modern-Era Retrospective Analysis for Research
61 and Applications, version 2 (MERRA2), and the National Centres for Environmental Prediction (NCEP) Final Operational

62 Global Analysis data (FNL). Although several studies have demonstrated that meteorological impacts derived from CTM
63 results can corroborate the results of TSM (Liu et al., 2023; Yan et al., 2024) or MLM (Ni et al., 2024; Yin et al., 2021),
64 uncertainties in the determination of meteorological effects on surface O₃ concentrations cannot be neglected. For example,
65 Pan et al. (2023) reported that the meteorological impact on O₃ trends in Beijing during 2013–2020 was +0.52 ppb yr⁻¹, which
66 is only half of the value estimated by Gong et al. (2022).

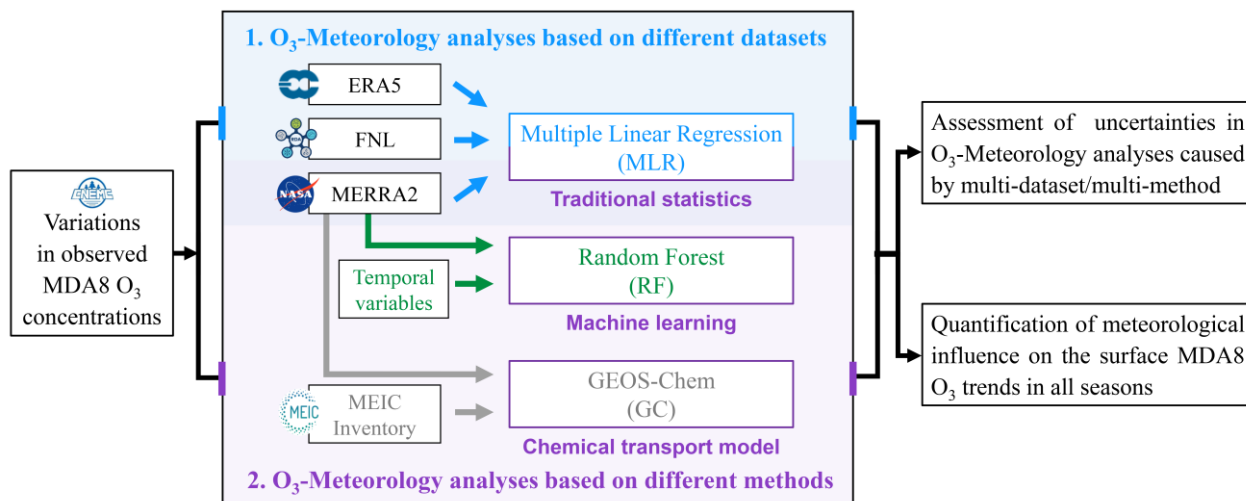
67 Uncertainties in quantifying the drivers of O₃ trends can be ascribed to the discrepancies between different meteorological
68 datasets and between different methods (Guo et al., 2021; Weng et al., 2022; Yao et al., 2024). Regarding the uncertainty
69 caused by different meteorological datasets, the meteorologically driven annual variations of O₃ concentrations from 2017 to
70 2019 identified by the MERRA2-driven TSM are not consistent with the ERA5-driven TSM during the summer of YRD (Hu
71 et al., 2024a; Qian et al., 2022). During the summer of 2013–2019 in YRD, Li et al. (2019) reported a trend of +0.7 ppb yr⁻¹
72 in meteorology-driven O₃ concentrations using the MERRA2-driven TSM, while the trend of Yan et al. (2024) was –0.3 µg
73 m⁻³ yr⁻¹ using the ERA5-driven TSM. Regarding the uncertainty caused by different methods, the meteorology-driven O₃ trend
74 identified by MLM for 2019–2021 was 2.4 times larger than that identified by CTM based on the same meteorological dataset
75 input (MERRA2) in the North China Plain (NCP) during summer (Wang et al., 2024a). In BTH, from 2021 to 2022, Luo et al.
76 (2024) identified a negative meteorological contribution based on the ERA5-driven MLM, while Yan et al. (2024) suggested
77 a positive contribution (+4.3 µg m⁻³) based on the ERA5-driven TSM during summer.

78 On the basis of the above-mentioned, large uncertainties caused by multi-dataset or multi-method exist in O₃-Meteorology
79 analyses. However, available intercomparisons of O₃ analyses mainly focused on predicting the O₃ concentrations. For
80 example, Wang et al. (2024b) and Weng et al. (2023) compared the differences in O₃ concentration prediction caused by
81 different datasets and models, respectively. The uncertainties in quantifying the meteorological contributions to O₃ trends
82 caused by multi-dataset and multi-method remain unassessed. In addition, previous studies have predominantly focused on
83 summer O₃ pollution, although reports indicate an extension of the O₃ pollution season to winter and spring across major
84 clusters in China (Cao et al., 2024a; Li et al., 2021) and an unfavourable meteorological impact on O₃ air quality in spring and
85 winter in BTH (Luo et al., 2024). It is essential to conduct an intercomparison of meteorology-driven O₃ quantification using
86 multi-dataset and multi-method across all seasons.

87 This study ~~utilized~~utilised 10-year (2013–2022) surface O₃ observations in China to investigate long-term O₃ trends and
88 quantify the meteorological influence on O₃ trends using diverse meteorological datasets and analytical methods. **Figure 1**
89 shows the framework and the main objectives were: (1) to assess uncertainties in identifying the meteorological influences
90 caused by multi-dataset. This was achieved by employing the TSM (i.e. multiple linear regression, MLR) driven by different
91 reanalysis meteorological products (i.e. ERA5, MERRA2, and FNL); (2) to assess uncertainties in identifying meteorological
92 effects caused by multi-method. This was achieved by establishing three models corresponding to TSM (i.e. MLR), MLM (i.e.
93 random forest, RF), and CTM (i.e. GEOS-Chem, GC), each driven by the MERRA2 product; (3) to calculate the mean of

94 meteorology-driven O₃ trends driven by three datasets (multi-dataset mean) and three methods (multi-method mean) to derive
 95 relatively robust results.

96 Our paper is structured as follows: Section 2 briefly introduces the details of surface O₃ observations and different
 97 meteorological datasets, as well as the framework of three methods, namely MLR, RF, and GC. The quantification of the
 98 uncertainties in the meteorology-driven O₃ trends caused by multi-dataset and multi-method is presented in Section 3. Section
 99 4 concludes the paper. The findings of this study provide a scientific foundation for developing regional and seasonal strategies
 100 to mitigate and manage O₃ pollution in China.



101
 102 **Figure 1.** The framework of the uncertainty assessment in this study.

103 2 Data and Methods

104 2.1 Surface O₃ and meteorological data sources

105 Hourly surface O₃ observations from over 1000 state-controlled stations operated by the China National Environmental
 106 Monitoring Centre from 2013 to 2022 were used to ~~analyze~~ analyse the long-term O₃ trends across all seasons: spring (March-
 107 April-May), summer (June-July-August), autumn (September-October-November), and winter (December-January-February).
 108 The maximum daily 8-hour average (MDA8) O₃ was calculated as an air quality indicator after filtering out abnormal data
 109 using the z-scores method. For detailed information on data quality control, refer to He et al. (2017).

110 In this study, we selected three widely used reanalysis products to assess the uncertainties caused by different meteorological
 111 datasets. Variables during 2013–2022 from ERA5, MERRA2, and FNL, as detailed in [Table S2](#), were selected as
 112 meteorological inputs for building MLR models. These reanalyses have spatial resolutions of 0.25°×0.25°, 0.625°×0.5°, and

113 $1^\circ \times 1^\circ$ on a global latitude-longitude grid, respectively. In Section 3.2, we have also incorporated the NCEP FNL reanalysis
114 product with a spatial resolution of $0.25^\circ \times 0.25^\circ$ (FNL025) for the period 2016–2022 to explore the effect of spatial resolution
115 on the analysis of uncertainties caused by multi-dataset.

116 2.2 Methods for obtaining long-term series and meteorological influence

117 2.2.1 Kolmogorov–Zurbenko (KZ) filter

118 The KZ filter, known for its ability to extract low frequency signals from time series data and handle missing values, has been
119 extensively applied in ~~analyzing~~ analysing air quality variations (Eskridge et al., 1997; Rao and Zurbenko, 1994; Wise and
120 Comrie, 2005). This filter is particularly useful in studying variations in air quality over time. The original time series of air
121 pollutants or meteorological variables ($X(t)$) can be decomposed by the KZ filter into the following form:

$$X(t) = X_{ST}(t) + X_{SN}(t) + X_{LT}(t) \quad (1)$$

$$X_{LT}(t) = KZ_{(365,3)}X(t) \quad (2)$$

$$X_{BL}(t) = KZ_{(15,5)}X(t) \quad (3)$$

$$X_{ST}(t) = X(t) - X_{BL}(t) \quad (4)$$

122 In the decomposition process, $X(t)$ represents the original daily time series, while $X_{ST}(t)$, $X_{SN}(t)$, and $X_{LT}(t)$ denote the
123 short-term, seasonal, and long-term components, respectively. The baseline component, $X_{BL}(t)$, is defined as the sum of
124 $X_{SN}(t)$ and $X_{LT}(t)$. The $KZ_{(p,q)}$ filter executes q iterations with p as the moving average window length of ~~the~~ $X(t)$ series.
125 Specially, ~~the~~ $X_{LT}(t)$ ~~series~~ is derived using the $KZ_{(365,3)}$ filter, capturing long-term changes with periods exceeding 1.7 years.
126 ~~The~~ $X_{BL}(t)$ ~~series~~ is obtained through the $KZ_{(15,5)}$ filter, encompassing both seasonal and long-term components, ~~while the~~
127 $X_{ST}(t)$ ~~series~~ represents short-term fluctuations with period less than 33 days in the original time series. $X_{SN}(t)$ is derived as
128 the difference between $X_{BL}(t)$ and $X_{LT}(t)$, corresponding to seasonal variation on a timescale of months. The KZ filter can
129 fill in missing values by using iterated moving average technique. Although not all of the ozone measurement sites were active
130 over the entire period 2013–2022, missing value problem can be handled for most stations after we conduct three iterations
131 with 365-day moving average.

132 In this study, all statistical analyses were performed at the seasonal scale (spring: March-April-May; summer: June-July-
133 August; autumn: September-October-November; winter: December-January-February). For each season, the $KZ_{(365,3)}$ filter
134 was applied to extract the long-term trends in observed, meteorology-driven, and emission-driven MDA8 O₃ concentrations
135 (see details in [Fig. S1](#)) during 2013–2022, as detailed in Sections 2.2.2, 2.2.3, and 2.2.4.

136 2.2.2 Stepwise MLR for separating meteorological influence

137 As vividly illustrated in [Fig. S1](#), a data-based TSM (i.e., MLR integrating the KZ filter) was employed to separate the observed
138 MDA8 O₃ concentrations into meteorology-driven and emission-driven concentrations (Sadeghi et al., 2022; Shang et al., 2023;

139 Zhang et al., 2022a). We initially applied the KZ filter to disassemble the MDA8 O₃ time series and all meteorological variables
140 listed in [Table S2](#) into short-term, baseline, and long-term components at individual state-controlled stations for each season.
141 Subsequently, a series of screening processes aligned with our previous research (Wang et al., 2024c), were executed to
142 perform stepwise MLR on the short-term/baseline MDA8 O₃ concentrations and a group of meteorological variables series,
143 respectively. The established MLR model is presented herein:

$$C_{s,r}(t) = b_{0,s,r} + \sum_{i=1}^k b_{i,s,r} \times Met_i(t) + \varepsilon \quad (5)$$

144 Here, $C_{s,r}(t)$ represents the MDA8 O₃ concentration for season s and monitoring station r , while $Met_i(t)$ signifies the i -th
145 meteorological variable out of a total of k , and $b_{i,s,r}$ is the corresponding regression coefficient. $b_{0,s,r}$ denotes the intercept
146 term, and ε is the residual term. After establishing MLR models for the short-term and baseline components in each season,
147 we obtain their respective residual terms. The total residuals, which represent the sum of residuals from baseline variables and
148 short-term variables, primarily reflect anthropogenic influences. We then applied a $KZ_{(365,3)}$ filter to these aggregated
149 residuals to derive long-term emission-driven and meteorology-driven O₃ variations. Finally, the meteorology-driven O₃ trends
150 and emission-driven O₃ trends were obtained through Least Square Method.

151 The constructed MLR models driven by meteorological variables from ERA5, MERRA2, or FNL in each season will ~~be~~
152 ~~constructed,~~ allow ing a comprehensive analysis of multi-dataset uncertainties. The meteorological impact on O₃ trends derived
153 from the MERRA2-driven MLR model will also be integrated into the analysis of multi-method uncertainties to improve the
154 comparability of results.

155 2.2.3 Random forest (RF) for deriving meteorological influence

156 The application of MLM in O₃ air quality research is becoming increasingly prevalent due to its superior accuracy, user-
157 friendly nature, and capability to capture nonlinear relationships (Ni et al., 2024; Yao et al., 2024; Zhang et al., 2022b).
158 Considering the limited influence of discrepancy in O₃-Meteorology analyses stemming from different machine learning
159 algorithms (Wang et al., 2024a), we opted to build a representative MLM known as the meteorological normalisation model
160 based on the RF algorithm (Ding et al., 2023; Ji et al., 2024; Zhang et al., 2023), to delineate meteorology- and emission-
161 driven O₃ concentrations.

162 RF stands out as a tree-based ensemble learning algorithm adept at handling nonlinear issues and reducing overfitting (Breiman,
163 2001). An RF model was developed for each state-controlled station in each season to predict the MDA8 O₃ concentration
164 using the Python package “Sklearn-RandomForestRegressor”. The predictors included six temporal variables (year, month of
165 a year, day of a week, day of a month, day of a year, Unix time), serving as proxies for anthropogenic emission intensity
166 (Grange et al., 2018), alongside six MERRA2 meteorological variables as listed in [Table S2](#) (i.e. SLP, T2max, U10, V10,
167 RH2, PBLHday). The training dataset comprised 70% of the data, while the remaining 30% was reserved for model evaluation.
168 A statistical cross-validation technique was employed to determine optimal hyperparameters for enhancing RF prediction

performance (Weng et al., 2022). Coefficient of determination (R^2) values were ~~utilized~~utilised to assess model performance for each station, ~~with stations exhibiting $R^2 < 0.5$ (marked in blue in Fig. S2b) being excluded to ensure model credibility.~~ Over 70% of state-controlled stations showed $R^2 > 0.5$ in all seasons (Fig. S2b), which is consistent with the 0.4–0.6 range reported in comparable studies (Weng et al., 2022; Lu et al., 2024). Stations with $R^2 < 0.5$ were excluded to avoid significant attribution uncertainty that could be introduced by the RF performance.

After establishing the RF model, both the original time variables and resampled meteorological variables were ~~utilized~~utilised as input data. ~~The meteorological variables were resampled by randomly selecting data from the two weeks before and after the specified date. To derive the de-weathered MDA8 O_3 concentration for a given day (e.g., March 1, 2013), the random resampling process was iterated a thousand times. Our methodology closely follows that of Vu et al. (2019), with a more detailed process shown in Fig. S2(a). The meteorology driven MDA8 O_3 concentrations were computed as the difference between the observed concentrations and de-weathered concentrations (i.e. emission driven MDA8 O_3 concentrations). For meteorological normalisation, we implemented the protocol of Vu et al. (2019). Meteorological variables were resampled by randomly selecting data from the two weeks before and after the specified date, while temporal proxies remained fixed. To derive the de-weathered MDA8 O_3 concentration for a given day (e.g. March 1, 2013), the random resampling process was iterated 1000 times. The mean predicted O_3 under average meteorological conditions, which refers to de-weathered O_3 , corresponds to the emission-driven O_3 concentration. The meteorology-driven MDA8 O_3 concentrations for each season were computed as the difference between observed concentrations and de-weathered concentrations. Detailed processes were shown in Fig. S2(a). The $KZ_{(365,3)}$ filter was then applied to obtain long-term components, and meteorology-driven O_3 trends were derived using Least Square Method.~~

2.2.4 GEOS-Chem (GC) simulation for quantifying meteorological influence

The numerical analysis of surface O_3 in China was performed with the GC classic version 13.3.3 (<https://github.com/geoschem/GCClassic/releases/tag/13.3.3>). Developed as a global 3-D model, GC incorporates a fully coupled O_3 –NO $_x$ –VOCs–aerosol–halogen chemical mechanism, driven by the MERRA2 meteorological input. Numerous studies have leveraged GC to simulate O_3 air quality in China, demonstrating alignment between observational data and model outcomes (Dai et al., 2024; Dang et al., 2021; Li et al., 2019; Lu et al., 2019a). We employed the nested-grid GC to simulate the long-term surface O_3 concentrations and to quantify the meteorology-driven MDA8 O_3 trends over China. The nested-grid domain was set over China’s mainland (15–55°N, 70–140°E) with a horizontal resolution of 0.5° latitude by 0.625° longitude and 47 vertical layers extending up to an altitude of 0.01 hPa. A global simulation with a horizontal resolution of 2°×2.5° provided the chemical boundary conditions for the nested-grid simulation every 3 hours. To ensure model stability and accuracy, a 6-month spin-up simulation was conducted before the commencement of the targeted 10-year period from March 2013 to February 2023.

200 Emissions management within GC is facilitated by the Harmonized Emissions Component, a system introduced by (Lin et al.,
201 Anthropogenic emissions are sourced from the Community Emissions Data System (CEDs) inventory globally, with
202 specific overwriting by the Multi-resolution Emission Inventory for China (MEIC) within the Chinese region. The simulations
203 for 2021–2022 adopt a similar approach to Zhai et al. (2021), using 2019 MEIC emissions with NO_x emissions reduced by 8
204 ~ 13% and 2017 MEIC with VOCs emissions reduced by 10 ~ 14%, based on the policy released by Ministry of Ecology and
205 Environment of the People's Republic of China. For natural emissions, biogenic VOCs, soil, and lightning NO_x were calculated
206 online in the model. Emissions from biomass burning, ships, and aircraft are sourced from the Global Fire Emissions Database,
207 the CEDs inventory, and the 2019 Aircraft Emissions Inventory Code, respectively.

208 In order to assess the model's performance and to get a quantification of meteorology-driven O₃ trends during the period of
209 2013–2022, two sets of simulations were conducted: (1) BASE: the standard simulation of O₃ concentrations from 2013 to
210 2022, where both meteorological fields and emissions (including anthropogenic, natural, and biomass emissions) vary year by
211 year from 2013 to 2022; (2) FixE2013: a “fixed-emission simulation” where meteorological conditions vary from 2013 to
212 2022 while anthropogenic emissions remain constant at 2013 levels. The FixE2013 simulation is designed to quantify the
213 meteorological influence on O₃ variations. The FixE2013 simulation is designed to obtain the MDA8 O₃ concentrations driven
214 solely by meteorological changes and further quantify the meteorological influence on O₃ variations in four seasons. After
215 applying the KZ_(365,3) filter to derive the long-term meteorology-driven series, trends were calculated through Least Square
216 Method. Figure S3 evaluates the performance of the GC simulation for 2013–2022. The GC model generally captures the
217 monthly variability in MDA8 O₃ over China and three cluster megacities, with the correlation coefficients greater than 0.80,
218 although it always shows a high bias of surface O₃ in warm seasons (Dai et al., 2024), which can be attributed to its inability
219 to capture the complex terrain, local pollution sources and meteorological conditions, or overestimates of the correlations
220 between the surface O₃ concentration and temperature (Shen et al., 2022; Sun et al., 2021).

221 2.3 Assessment of uncertainties caused by multi-dataset and multi-method

222 In this study, the coefficient of variation (CV) is applied to assess the uncertainties in O₃-Meteorology analyses caused by
223 different meteorological datasets or methods. The CV, calculated as the ratio of the standard deviation (SD) to the mean, serves
224 as a statistical metric commonly ~~utilized~~ utilised to measure the diversity within datasets or models (Bedeian and Mossholder,
225 2000; Chen et al., 2019). Compared to other comparators (e.g. range, inter-quartile range, and SD), the CV is a unit-free
226 measure that quantifies percentage variation relative to the mean and is less sensitive to outliers and heavy-tailed distributions
227 (Högel et al., 1994; Chattamvelli and Shanmugam, 2023). In this study, higher CVs indicate lower consistency of
228 meteorologically driven O₃ trends derived from different datasets or methods. To give a more quantitative assessment,
229 consistency levels were classified as strong and weak with CV<0.5 and CV>1.0, respectively (Wang et al., 2022a). Given the
230 possibility of disparate meteorology-driven O₃ trends detected by different datasets or methods, we consider the absolute value
231 of the CV as a quantitative indicator of the uncertainties. For each season, When-when examining the uncertainties arising

from different datasets, the CV represents the ~~standard deviation~~SD of trends derived from the ERA5, MERRA2, and FNL-driven MLR models divided by the mean. Similarly, in the context of multi-method uncertainties, the CV is the ~~standard deviation~~SD of trends identified by the MLR, RF, and GC models divided by the mean.

3 Results

3.1 Observed trends in surface O₃ concentration

Figure 2 shows the trends in observed MDA8 O₃ concentrations over a 10-year period during four seasons. Noteworthy increases in O₃ concentrations were observed at 78 ~ 93% of state-controlled stations over the years, with the national trend being +1.31 ppb yr⁻¹, +0.93 ppb yr⁻¹, +0.79 ppb yr⁻¹, and +0.80 ppb yr⁻¹ in spring, summer, autumn, and winter, respectively.

The major eastern megacity clusters in China also displayed their highest MDA8 O₃ increase trends in spring, with trends of +1.16 ppb yr⁻¹ in BTH, +1.61 ppb yr⁻¹ in YRD, and +1.48 ppb yr⁻¹ in PRD, which has been reported in previous studies (Cao et al., 2024b; Chen et al., 2020; Wang et al., 2022**b**). During summer, BTH and YRD faced more severe challenges in O₃ prevention and control compared to PRD, with rising MDA8 O₃ trends in the former two regions being about three times higher than that in PRD (**Fig. 2b**).

In terms of O₃ growth rates, Shanxi province and Anhui province ranked the top two provinces in China over the past decade in all seasons except for winter, consistent with Zhao et al. (2020). In spring and winter, O₃ concentrations increased in all provinces, with trends of +0.39 ~ +2.75 ppb yr⁻¹ and +0.42 ~ +1.30 ppb yr⁻¹, respectively. Notably, Jilin province experienced an obvious improvement in O₃ air quality during summer and autumn, with decreasing trends of -0.74 ppb yr⁻¹ and -0.38 ppb yr⁻¹, respectively, which was also confirmed by Gong et al. (2022). As mentioned in Section 1, variations in O₃ concentrations are fundamentally modulated by emissions and meteorology. This section mainly documents observed O₃ trends, and the quantitative contributions of emissions and meteorology to MDA8 O₃ variations will be discussed in Section 3.2.

The annual and seasonal mean MDA8 O₃ concentrations across China are detailed in **Fig. S4** and **Fig. S5**, providing a holistic depiction of the persisting spread of O₃ pollution since 2013. On a national average, the O₃ air quality was worst in summer, with the average O₃ levels exceeding the air quality standard Grade I limit of 50 ppb almost every year. Notably, the summer of 2019 marked a peak period for O₃ pollution, with an average concentration of 59.7 ppb (**Fig. S5b**).

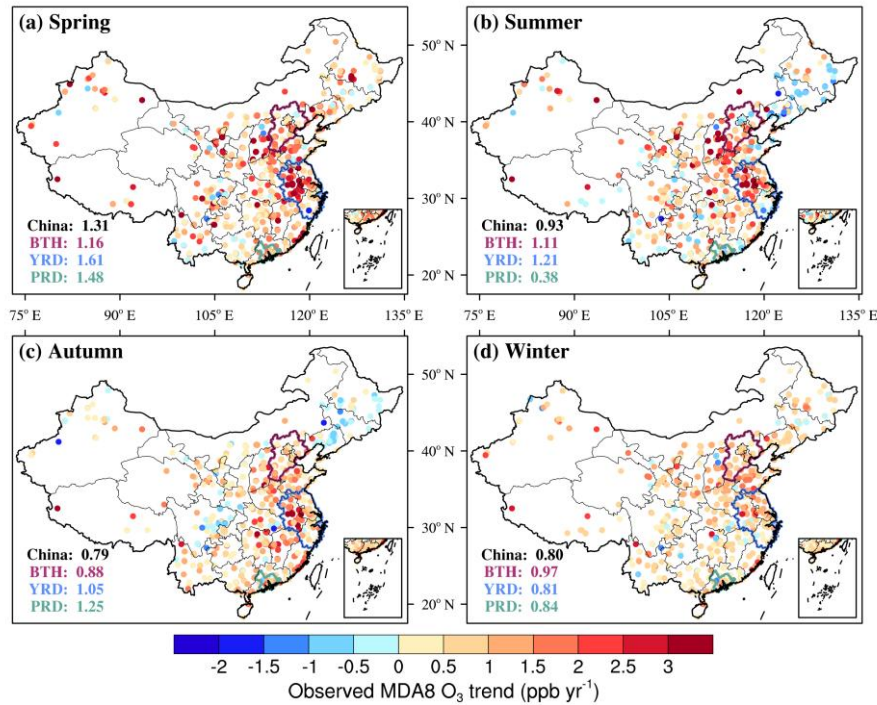


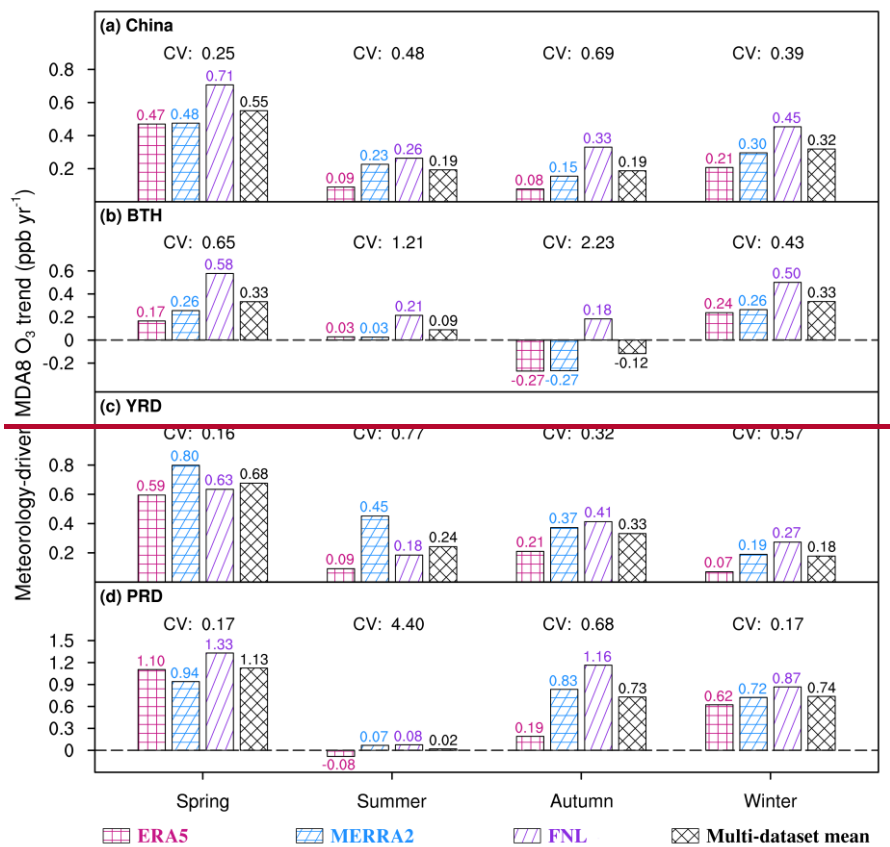
Figure 2. Trends in observed MDA8 O₃ concentrations in China from 2013 to 2022 during (a) spring, (b) summer, (c) autumn, and (d) winter. Values in black, purple, blue, and green represent the mean trends for the whole China, BTH, YRD, and PRD, respectively.

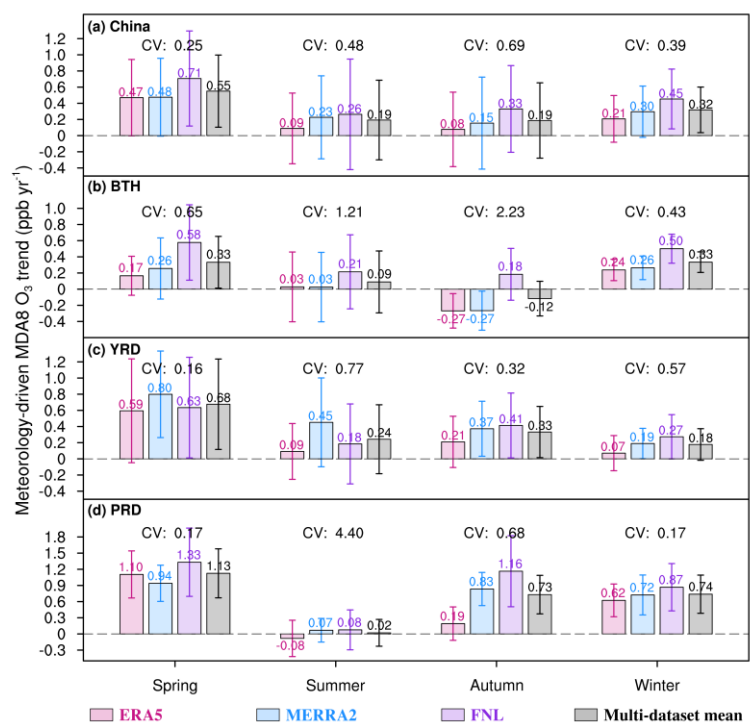
3.2 Uncertainty in meteorology-driven O₃ trends caused by multi-dataset

The traditional statistical method (the MLR model), which has a relatively low computational cost but can provide valuable insights into the quantification of meteorological contributions to O₃ trends, was used to investigate the uncertainties in O₃-Meteorology analyses caused by different meteorological datasets. As shown in [Fig. 3\(a\)](#), meteorological conditions contribute to an increase in MDA8 O₃ concentrations across all seasons in China, with the multi-dataset mean trends ranging from +0.19 (± 0.47) ppb yr⁻¹ to +0.55 (± 0.45) ppb yr⁻¹. All three dataset-driven MLR models indicate that meteorology leads to the most rapid increase in MDA8 O₃ concentrations in spring, with trends ranging from +0.47 (± 0.47) ppb yr⁻¹ to +0.71 (± 0.59) ppb yr⁻¹, and a low CV of 0.25. This suggests a high consistency among the three datasets in assessing the meteorological influence on surface O₃ concentrations. During summer and autumn, meteorological influences on O₃ show the greater spatial heterogeneity (with higher SD) and larger variability among multi-datasets (with higher CV). relatively higher CVs were shown, indicating larger variability, despite lower meteorological influences on O₃ trends. Specifically in autumn, the meteorology-driven O₃ trend derived from the FNL-driven MLR model is 4.1 times larger than that derived from the ERA5-driven MLR model. Lu et al. (2024) compared meteorology-driven O₃ trends derived from ERA5- and MERRA2-driven MLR models during the summer of 2013–2019. Their findings revealed that ERA5-derived trends were lower than those from MERRA2 in

273 YRD and PRD, whereas trends derived from ERA5 were comparable to those from MERRA2 in BTH. This inter-study
274 consensus further validates the robustness of our methodological framework.

275 **Figure 3(b-d)** depicts the meteorological impact on the MDA8 O₃ trends in the three megacity clusters (BTH, YRD, and PRD).
276 Meteorology caused the MDA8 O₃ increase in most of the megacity clusters and seasons, except for BTH during autumn. In
277 seasons where the meteorological effects derived from the three MLR models are all positive, the multi-dataset mean trends
278 ranged from +0.09 (±0.38) to +0.33 (±0.13) ppb yr⁻¹ in BTH, +0.18 (±0.20) to +0.68 (±0.56) ppb yr⁻¹ in YRD, and
279 +0.73 (±0.36) to +1.13 (±0.45) ppb yr⁻¹ in PRD. Consistent with **Fig. 3(a)**, meteorology triggered the most rapid increase
280 in MDA8 O₃ concentrations in spring across the three megacity clusters. The largest meteorological impact in BTH during
281 spring was also revealed by Luo et al. (2024). Large CVs (>1.0) were observed in BTH during summer and autumn. Notably,
282 the meteorological influence calculated by the three dataset-driven MLR models even showed opposite trends in BTH during
283 autumn, indicating challenges in assessing the meteorological impacts on surface O₃ concentrations. In contrast, in YRD and
284 PRD, the three MLR models demonstrated high consistency across almost all seasons. Although the largest CV reached 4.4 in
285 PRD during summer, it was considered acceptable because the three MLR models indicated that meteorology had a minor
286 influence (less than +0.1 ppb yr⁻¹) on O₃ trends.



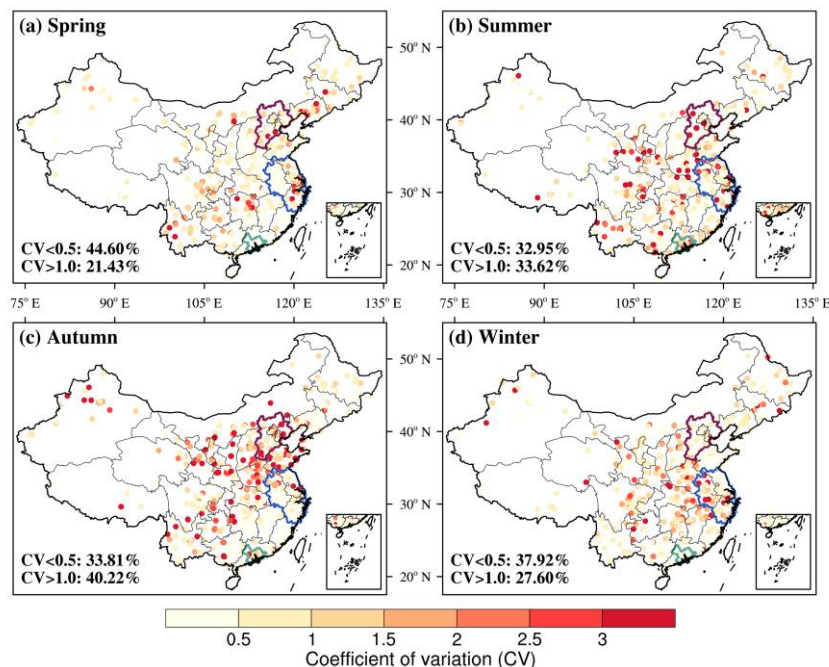


288

289 **Figure 3.** Meteorology-driven MDA8 O₃ trends in (a) the whole China, (b) BTH, (c) YRD, and (d) PRD during four seasons. Values in red,
 290 blue, and purple represent trends calculated by ERA5-, MERRA2-, and FNL-driven multiple linear regression (MLR) model, respectively.
 291 The fourth black bar represents the multi-dataset mean trend. Error bars indicate ±1 standard deviation (SD) of site-level trends calculated
 292 from all available monitoring stations within each region. The absolute value of the coefficient of variation (CV) for each season is also
 293 shown.

294 From a provincial perspective in [Fig. S6](#), we can also see that the meteorological contributions to O₃ trends are positive during
 295 spring and winter. Large uncertainties in O₃-Meteorology analyses were identified during summer and autumn. There were 7
 296 and 12 provinces with controversial meteorological contributions identified by the three dataset-driven MLR models in
 297 summer and autumn, respectively.

298 **Figure 4** displays the spatial distribution of the CV values from the perspective of state-controlled stations in four seasons.
 299 Consistent with the national and provincial perspectives, the least uncertainties in O₃-Meteorology analyses were observed in
 300 spring, with CVs less than 0.5 at 45% of stations. Obvious discrepancies in meteorology-driven O₃ trends are found in summer
 301 and autumn, particularly in the NCP and northwestern China, with CVs greater than 1.0 at 33 ~ 40% of the stations. In autumn,
 302 it is noteworthy that the uncertainties caused by multi-dataset are lower in the south than in the north. In a study using
 303 the Previous studies that employed MLR model to predict O₃ concentration, it was also revealed found that the MLR had
 304 better performance in the south than in the north (Han et al., 2020; Lu et al., 2024).



305

306 **Figure 4.** The absolute value of the coefficient of variation (CV) for each state-controlled monitoring station in China during (a) spring, (b)
 307 summer, (c) autumn, and (d) winter. The CV is calculated by the standard deviation (SD) of the trends derived from ERA5-, MERRA2-, and
 308 FNL-driven MLR models divided by the mean. The darker colour means the larger uncertainty in quantifying the meteorological impact on
 309 observed O₃ trends. The proportion of state-control stations with CV less than 0.5 and greater than 1.0 is also shown. The outline marked in
 310 purple, blue, and green represents the region of BTH, YRD, and PRD, respectively.

311 Based on the three dataset-driven MLR models, the meteorological and anthropogenic contributions to the MDA8 O₃ trends
 312 in China during 2013–2022 were further examined. As presented in [Fig. 5](#), both meteorological conditions and anthropogenic
 313 emissions lead to O₃ increases. According to the ERA5- and MERRA2-driven MLR models, variations in anthropogenic
 314 emissions were identified as the dominant factor driving the increase in MDA8 O₃ concentrations across all seasons, with
 315 anthropogenic contributions ranging from 63.2% to 90.4%. The results suggest that more stringent emission control policies
 316 should be implemented to counteract the adverse effects of meteorological influences on O₃ concentrations.

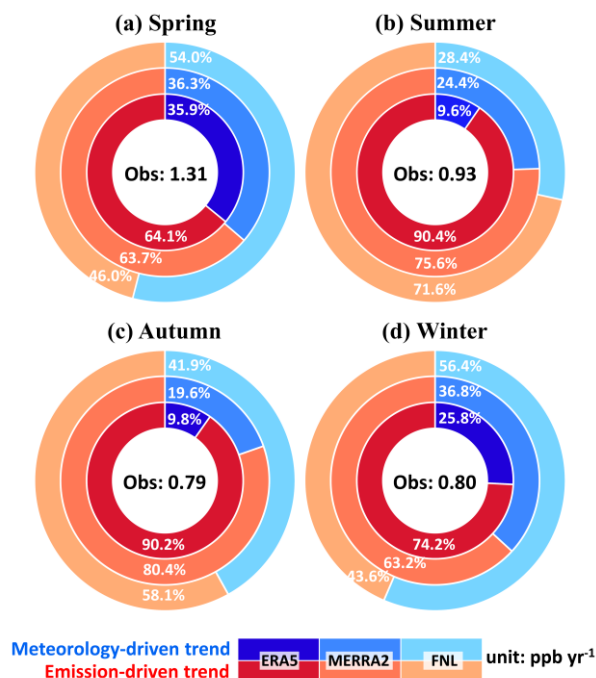
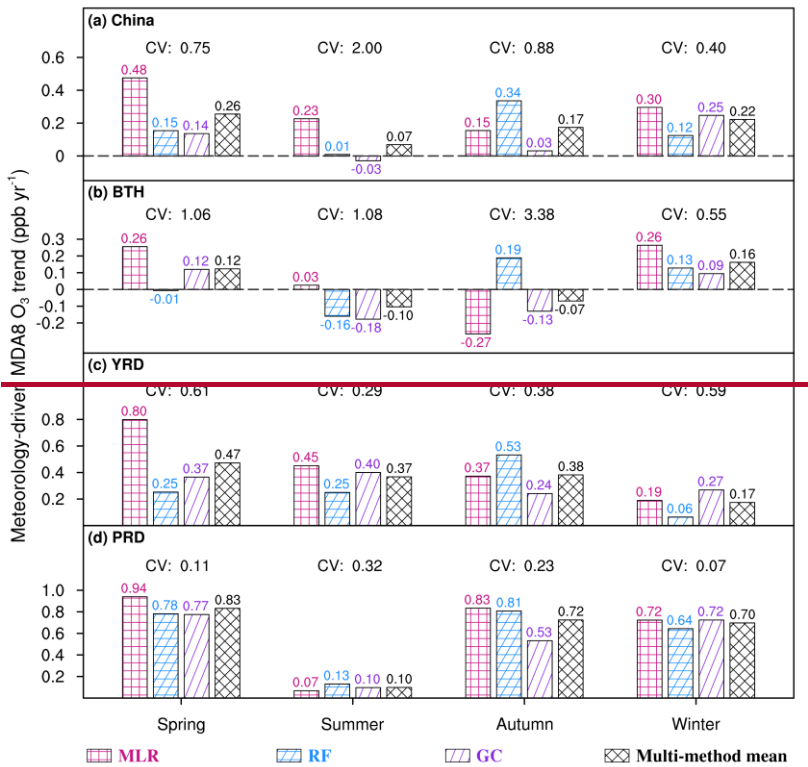


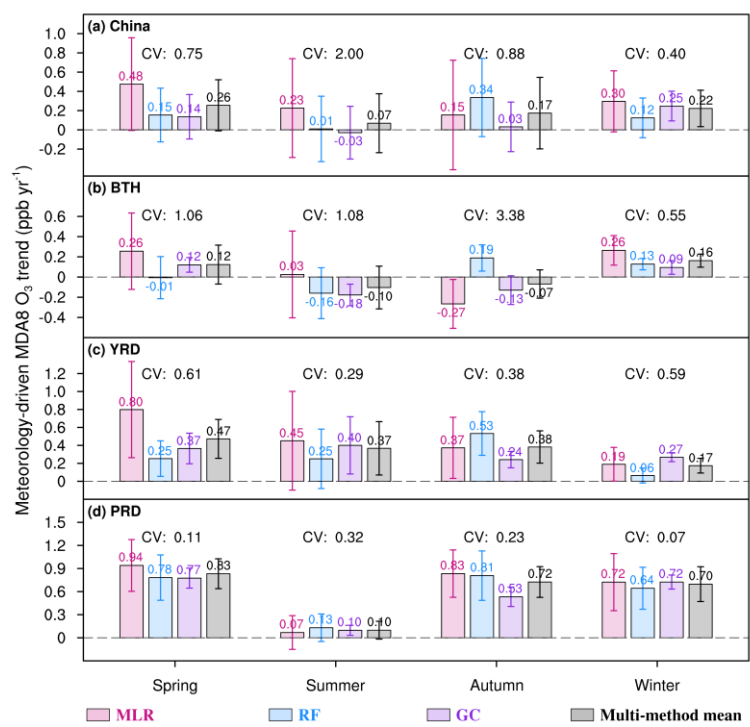
Figure 5. Percentage contributions of meteorological conditions (blue) and anthropogenic emissions (red) to the trends in observed MDA8 O₃ concentrations calculated by ERA5-, MERRA2-, and FNL-driven multiple linear regression (MLR) model in China during (a) spring, (b) summer, (c) autumn, and (d) winter. Values in black represent the observed MDA8 O₃ trends averaged over the whole China.

It is interesting to note that the FNL-driven model almost always gave relatively larger predictions of meteorologically driven O₃ trends compared to the models driven by ERA5 and MERRA2. To investigate whether this discrepancy was due to the coarser spatial resolution of the FNL dataset, a comparison was made between the FNL025-driven MLR model (0.25°×0.25°) and the FNL-driven MLR model (1.0°×1.0°). As depicted in Fig. S7, the deviation of the meteorology-driven trends calculated by the two MLR models was less than 0.1 in China and three megacity clusters across four seasons, indicating that different spatial resolutions have little effect on O₃-Meteorology analyses. Further examination was conducted to assess the influence of meteorological variables on O₃-Meteorology analyses. Table S3 lists the 10-year trends in each meteorological factor and shows a great discrepancy in the variable “PBLHday”. Zuo et al. (2023) also reported that FNL exhibited the highest uncertainty for the evaluation of PBLH compared to ERA5 and MERRA2, and that its performance may be constrained by complex underlying terrain and static instability (Guo et al., 2021). As Fig. S8 shows, constructing the FNL-driven MLR models using six meteorological variables without “PBLHday” can reduce the estimated meteorological impact by 0.08 to 0.20 ppb yr⁻¹. To obtain a more reliable estimate, it is recommended to use MERRA2 reanalysis dataset due to its eclectic result (Fig. 3) and avoid using FNL because of the uncertainty brought by PBLH “PBLH” to build the FNL-driven MLR models when separating meteorological and anthropogenic influences on O₃ concentrations in China.

335 **3.3 Uncertainty in meteorology-driven O₃ trends caused by multi-method**

336 This section discusses the uncertainties caused by multi-method (i.e. MLR, RF, GC), all of which are driven by the MERRA2
337 dataset. **Figure 6** illustrates the meteorology-driven MDA8 O₃ trends calculated by the MLR, RF, and GC models. For the
338 whole of China, the large uncertainties were evident during summer, when the meteorology-driven O₃ trends derived from the
339 MLR model are notably larger than those from the RF and GC models, with a CV of 2.0 (**Fig. 6a**). In the other three seasons,
340 ~~although~~ the multi-method mean trends, ranging from +0.17 (± 0.37) to +0.26 (± 0.27) ppb yr⁻¹, are 1.1 to 2.1 times lower
341 than those computed by the three dataset-driven MLR models (**Fig. 3a**), all models converge on the conclusion that
342 meteorological conditions contribute to the deterioration of O₃ air quality.





344

345 **Figure 6.** Meteorology-driven MDA8 O₃ trends in (a) the whole China, (b) BTH, (c) YRD, and (d) PRD during four seasons. Values in red,
 346 blue, and purple represent trends calculated by multiple linear regression (MLR), random forest (RF), and GEOS-Chem (GC) models,
 347 respectively. The fourth black bar represents the multi-method mean trend. Error bars indicate ±1 standard deviation (SD) of site-level trends
 348 calculated from all available monitoring stations within each region. The absolute value of the coefficient of variation (CV) for each season
 349 is also shown.

350 In YRD and PRD, the three models exhibit strong agreement in all seasons, with the largest CV of 0.61, where meteorology
 351 leads to an increase in O₃ concentrations with multi-method mean trends of +0.17 (±0.08) to +0.47 (±0.22) ppb yr⁻¹ and
 352 +0.10 (±0.12) to +0.83 (±0.19) ppb yr⁻¹, respectively. Notably, the most rapid meteorology-driven O₃ increase is also
 353 observed in spring (**Fig. 6c and Fig. 6d**), which is consistent with **Fig. 3c and Fig. 3d**. Lu et al. (2024) also demonstrated a
 354 high degree of consistency among the MLR, ML, and GC models in PRD during summer. Specifically, all three models
 355 indicated that meteorology contributed approximately 25% of O₃ variability over the period 2013–2019. In BTH, the three
 356 models perform consistently well only in winter, with meteorology-driven O₃ trends ranging from +0.09 (±0.07) to +0.26 (±
 357 0.15) ppb yr⁻¹ and a CV of 0.55. It is also observed that in summer and autumn, meteorology plays a relatively small role in
 358 influencing O₃ air quality despite the controversial results obtained by the three models (**Fig. 6b**). This finding aligns with a
 359 study focusing on the O₃ air quality in BTH from 2015 to 2022 (Luo et al., 2024), which suggested that meteorological
 360 conditions tend to increase MDA8 O₃ concentration by only 0.01 μg m⁻³ in summer and decrease MDA8 O₃ concentration by
 361 0.3 μg m⁻³ in autumn from 2015 to 2022.

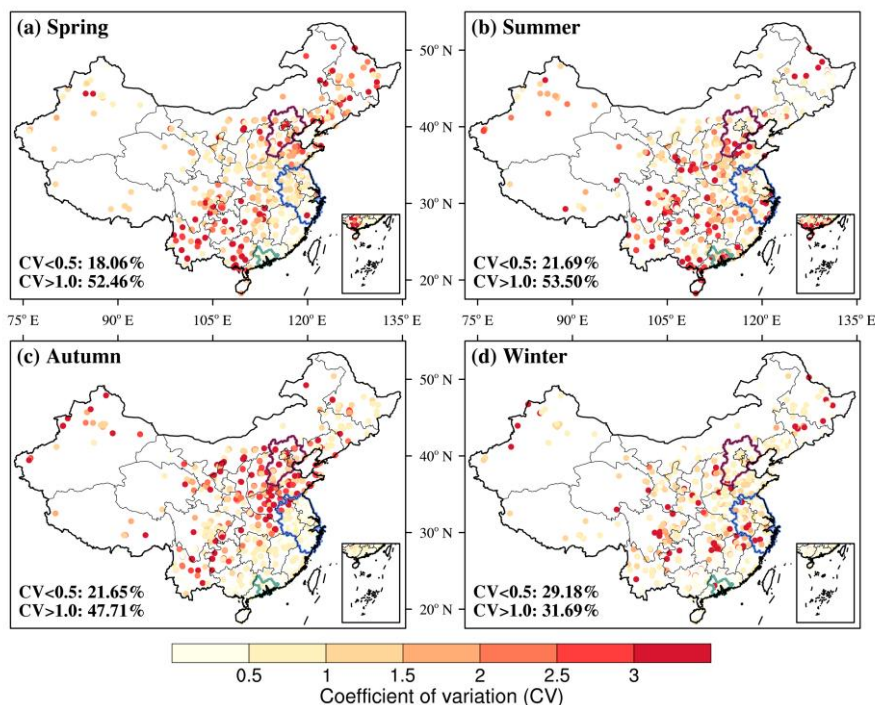


Figure 7. The absolute value of the coefficient of variation (CV) for each state-controlled monitoring station in China during (a) spring, (b) summer, (c) autumn, and (d) winter. The CV is calculated by the standard deviation (SD) of the trends derived from multiple linear regression (MLR), random forest (RF), and GEOS-Chem (GC) models divided by the mean. The darker colour means the larger uncertainty in quantifying the meteorological impact on observed O₃ trends. The proportion of state-control stations with CV less than 0.5 and greater than 1.0 is also presented. The outline marked in purple, blue, and green represents the region of BTH, YRD, and PRD, respectively.

In addition, Fig. 6 illustrates that the meteorology-driven O₃ trends obtained from GC are relatively smaller. As shown in Fig. S3 and Table S4, this difference could partly be attributed to the higher O₃ values-levels and lower O₃ increases simulated by the GC model before 2018. The GC's systematic overestimation of O₃ concentrations, as well as underestimation of O₃ increases, were also reported by Lu et al. (2024), in which the GC captured 13.6 ~ 81.1% of the observed O₃ increases in China during the summer of 2000–2019. It is crucial to take into account the overestimation of low-level O₃ observations, as noted in previous studies (Hu et al., 2024c; Mao et al., 2024). To validate this hypothesis, we compared the meteorology-driven O₃ trends calculated by MLR with those calculated by GC from 2018 to 2022, and a higher agreement was found over 2018–2022 compared to the 2013–2022 period in Fig. S9. The trends driven by RF model are eclectic in more cases (Fig. 6) and recommended to isolate meteorological and anthropogenic drivers.

From a provincial perspective, as depicted in Fig. S10, the three models together indicate that meteorology causes an O₃ increase in winter across almost all provinces except for Guizhou and Sichuan. In summer and autumn, meteorology leads to a decrease in 5 provinces, mainly in northeastern China, with trends ranging from –0.42 to –0.11 ppb yr^{–1}. Interestingly, across

all seasons, the three models introduce less uncertainty in the developed east coast regions such as Jiangsu, Fujian, and Guangdong compared to other provinces. This suggests that quantifying the impact of meteorological impact on O₃ levels in these developed regions along the east coast of China is relatively reliable.

From the perspective of state-controlled stations, **Fig. 7** shows the spatial distribution of the CV during four seasons. The lowest disparities in the meteorology-driven MDA8 O₃ trends persist in winter, with CVs of less than 0.5 recorded at 29% of the stations. In the other three seasons, however, significant discrepancies in meteorology-driven O₃ trends are prominent, with a CV greater than 1.0 at least 48% of the stations. Similar to **Fig. 4**, it is noteworthy that in autumn, the uncertainties caused by multi-method are more pronounced in the northern regions compared to the southern regions.

4 Limitations

While this study advances understanding of meteorological contributions to O₃ trends, several limitations warrant attention in future work. Though the reanalysis meteorological dataset is generated observationally, inherent constraints exist, including parameterization uncertainties affecting O₃-relevant physical processes (Janjić et al., 2018; Davidson and Millstein, 2022) and resolution constraints.

Regarding analytical approaches, machine learning efficiently captures nonlinear O₃-meteorology relationships without requiring explicit physicochemical parameterizations, enabling scalable multi-site analysis. However, its inability to resolve chemical mechanisms and sensitivity to predictor selection remain key constraints. Conversely, while GEOS-Chem mechanistically resolves chemistry-transport interactions and enables source attribution, it propagates uncertainties from emission inventories and chemical mechanisms into trend estimates.

Future studies could be improved in the following ways: First, more meteorological datasets and methods should be used to provide more robust uncertainty quantification in O₃-meteorology analyses. Second, implementing clustering techniques (e.g. K-means algorithm) could identify sub-regional drivers at ecotones, enhancing spatial resolution beyond our regional framework. Finally, the Lindeman-Merenda-Gold indices can be employed to quantitatively resolve the contributions of specific meteorological variables. The mechanistic understanding of O₃ drivers would be improved by integrating additional variables, such as solar radiation, soil moisture, and climate indices (e.g. El Niño-Southern Oscillation). Clustering techniques would be valuable to augment the region-based approach and would provide better understanding of the similarity between stations.

4.5 Conclusions and Discussions

This study used the 10-year (2013–2022) surface O₃ observations to clarify O₃ variations during four seasons in China, and quantify the meteorological impacts on O₃ trends, with a special focus on the uncertainties of meteorology-driven O₃ trends. Diverse meteorological datasets (ERA5, MERRA2, FNL) and analytical methods (MLR, RF, GEOS-Chem) were employed to systematically analyze the uncertainties in meteorology-driven O₃ trends caused by multi-dataset and multi-method which have not been assessed before. The coefficient of variation (CV) was adopted as a metric to assess the uncertainty. The main conclusions are as follows:

Over the past decade, increasing trends in MDA8 O₃ were observed at over 78% of state-controlled stations across all seasons, with the national trend of +1.31 ppb yr⁻¹, +0.93 ppb yr⁻¹, +0.79 ppb yr⁻¹, and +0.80 ppb yr⁻¹ in spring, summer, autumn, and winter, respectively.

We first applied the MLR model (driven by ERA5, MERRA2, and FNL, respectively), which has proven its usefulness and reliability in O₃-Meteorology analyses, to assess uncertainties caused by multi-dataset. For the whole China, all three dataset-driven MLR models indicate that meteorological conditions have led to an increase in MDA8 O₃ concentrations in four seasons, with multi-dataset mean trends ranging from +0.19 ppb yr⁻¹ to +0.55 ppb yr⁻¹. The models driven by different meteorological datasets showed a maximum meteorology-driven O₃ trend of +0.55 ppb yr⁻¹ with the highest consistency (CV=0.25) in spring. The FNL-driven model always obtained larger meteorology-driven O₃ trends compared to the models driven by ERA5 and MERRA2, which could be attributed to the inability to accurately evaluate PBLH in the FNL dataset. The dominant influence of anthropogenic emissions on O₃ increase was also identified, highlighting the need for more stringent emission control policies to mitigate the adverse effects of meteorological conditions.

We further applied the MLR, RF and GEOS-Chem model to obtain the meteorological influence on O₃ trends to explore the uncertainties caused by multi-method. In China and three megacity clusters, the three methods consistently indicated positive meteorological contributions to O₃ increases during spring and winter, with multi-method mean trends ranging from +0.12 to +0.83 ppb yr⁻¹ and +0.17 to +0.70 ppb yr⁻¹, respectively. In summer and autumn, especially in BTH, where the meteorological influence was relatively lower, three methods gave conflicting predictions of meteorological influence on O₃ with CVs greater than 1.08. For the whole China, three different methods demonstrated optimal consistency in winter with CV of 0.40 and the worst consistency in summer with CV of 2.00. The meteorology-driven O₃ trends obtained from GEOS-Chem model were almost relatively smaller than those obtained by other two methods, which could partly be attributed to the higher O₃ values simulated by the GEOS-Chem model before 2018.

All analyses driven by diverse meteorological datasets and analytical methods drew a consistent finding: meteorological conditions almost contribute to O₃ increase across all seasons. The uncertainties of meteorology-driven O₃ trends caused by different analytical methods were larger than those caused by diverse meteorological datasets.

437 ~~While this study advances understanding of meteorological contributions to O₃ trends, there exist several limitations that~~
438 ~~should be overcome in future studies. More meteorological datasets and methods should be used to provide a more accurate~~
439 ~~assessment of uncertainties in O₃ Meteorology analyses.~~ Considering that the favourable effects of meteorology on O₃ pollution
440 tend to be weaker after 2019 and the effects of COVID-19, it is necessary to conduct research over different periods and longer
441 periods. In addition, further research is needed to focus on the meteorological contributions to O₃ trends in northern China due
442 to larger uncertainties.

443 **Data availability.** The surface O₃ observations are obtained from <https://quotsoft.net/air/>. The ERA5, MERRA2, FNL, and
444 FNL025 reanalysis meteorological data are taken from <https://cds.climate.copernicus.eu/datasets>,
445 http://geoschemdata.wustl.edu/ExtData/GEOS_0.5x0.625_AS/MERRA-2/, <https://rda.ucar.edu/datasets/d083002/>, and
446 <https://rda.ucar.edu/datasets/ds083-3/>, respectively. The code of the GEOS-Chem (version 13.3.3) model is available at
447 <https://zenodo.org/records/5748260>. ~~The MDA8 O₃ observations and analytical results derived from MLR, RF, and GEOS-~~
448 ~~Chem can be obtained from <https://doi.org/10.5281/zenodo.15859028>.~~

449 **Author contributions.** JZ designed the research, and XW performed simulations and ~~analyzed~~ analysed the results. GJ, XC,
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452 **Competing interests.** The authors declare that they have no conflict of interest.

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