

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

Xueqing Wang¹, Jia Zhu^{1*}, Guanjie Jiao¹, Xi Chen¹, Zhenjiang Yang¹, Lei Chen^{1,2}, Xipeng Jin¹, and Hong Liao¹

¹Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Jiangsu Collaborative Innovation Centre of Atmospheric Environment and Equipment Technology, Joint International Research Laboratory of Climate and Environment Change, School of Environmental Science and Engineering, Nanjing University of Information Science and Technology, Nanjing, 210044, China

²State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex, Beijing, China

Correspondence: Jia Zhu (jiazhu@nuist.edu.cn)

Abstract. China has witnessed notable increases in surface ozone (O_3) 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 O_3 records (2013–2022) across China, revealing intensified nationwide O_3 pollution with increasing O_3 trends of 0.79–1.31 ppb yr^{-1} during four seasons. We gave special focus on uncertainties of meteorology-driven O_3 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 O_3 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 O_3 trends obtained from GEOS-Chem model were almost smaller than those obtained by other two methods, partly resulting from higher simulated O_3 values before 2018. Overall, all analyses driven by diverse meteorological datasets and analytical methods drew a robust conclusion that meteorological conditions almost boosted O_3 increases during all seasons; the uncertainties caused by different analytical methods were larger than those caused by diverse meteorological datasets.

Keywords: Meteorological impact, O_3 trend, Uncertainty

30 1 Introduction

31 Since 2013, the Chinese government has implemented a series of policies to mitigate air pollution resulting from the rapid
32 industrial and urban expansion, such as the “Air Pollution Prevention and Control Action Plan” (Wang, 2021). Several criteria
33 air pollutants exhibited decreases due to the emission control efforts, but not ozone (O₃) (Qi et al., 2023; Shen et al., 2020). In
34 China, O₃ concentrations were increased by 50–124 µg m⁻³ from 2015 to 2022 (Yao et al., 2024). The formation of surface O₃
35 depends nonlinearly on its precursors and is strongly influenced by meteorological conditions and anthropogenic emissions
36 (Wang et al., 2017). The impact of emission-related factors on O₃ increases in China over the past decade has been extensively
37 debated, including the ineffective control of volatile organic compounds (VOCs) emissions, the heightened O₃ photochemical
38 production due to the rapid decrease in PM_{2.5}, and the reduced nitric oxide (NO) titration effect (Li et al., 2019, 2022; Lin et
39 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 research focusing on longer time frames to gain deeper insights
55 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).

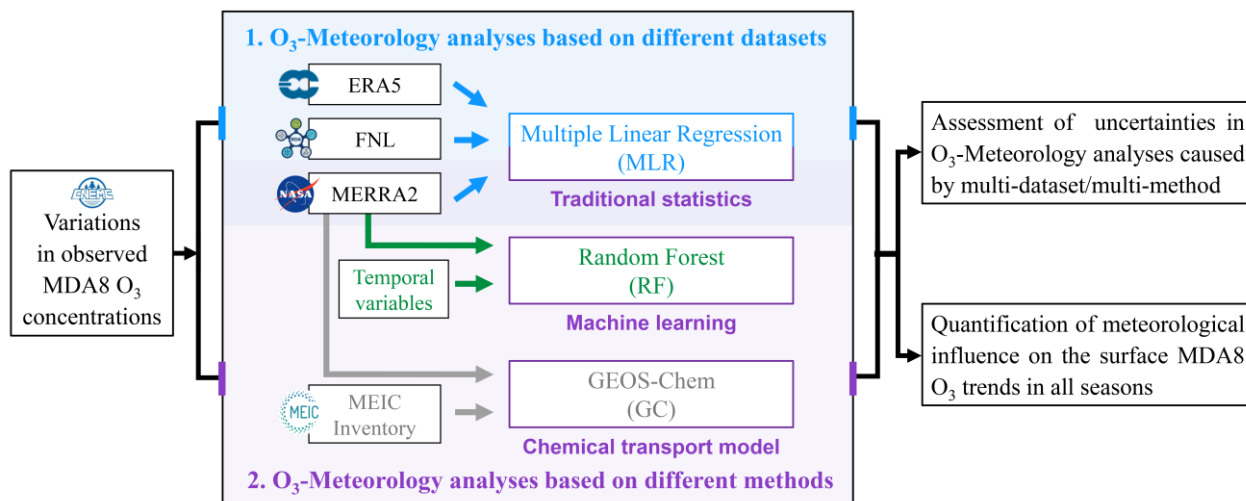
67 Uncertainties in quantifying the drivers of O₃ trends can be ascribed to the discrepancies between different meteorological
68 datasets and between different methods (Guo et al., 2021; Weng et al., 2022; Yao et al., 2024). Regarding the uncertainty
69 caused by different meteorological datasets, the 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 summers 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 utilised 10-year (2013–2022) surface O₃ observations in China to investigate long-term O₃ trends and quantify the
88 meteorological influence on O₃ trends using diverse meteorological datasets and analytical methods. **Figure 1** shows the
89 framework, and the main objectives were: (1) to assess uncertainties in identifying the meteorological influences caused by
90 multi-dataset. This was achieved by employing the TSM (i.e. multiple linear regression, MLR) driven by different reanalysis
91 meteorological products (i.e. ERA5, MERRA2, and FNL); (2) to assess uncertainties in identifying meteorological effects
92 caused by multi-method. This was achieved by establishing three models corresponding to TSM (i.e. MLR), MLM (i.e. random
93 forest, RF), and CTM (i.e. GEOS-Chem, GC), each driven by the MERRA2 product; (3) to calculate the mean of meteorology-

94 driven O₃ trends driven by three datasets (multi-dataset mean) and three methods (multi-method mean), so as to 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 meteorology-driven O₃ trends caused by multi-dataset and multi-method is presented in Section 3. Section 4
 99 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 analyse the long-term O₃ trends across all seasons: spring (March-April-
 107 May), summer (June-July-August), autumn (September-October-November), and winter (December-January-February). The
 108 maximum daily 8-hour average (MDA8) O₃ was calculated as an air quality indicator after filtering out abnormal data using
 109 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 also incorporate the NCEP FNL reanalysis product
 114 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 on the
 115 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 to analysing air quality variations (Eskridge et al., 1997; Rao and Zurbenko, 1994; Wise and Comrie,
 120 2005). This filter is particularly useful in studying variations in air quality over time. The original time series of air pollutants
 121 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 $X(t)$ series.
 125 Specially, $X_{LT}(t)$ is derived using the $KZ_{(365,3)}$ filter, capturing long-term changes with periods exceeding 1.7 years. $X_{BL}(t)$
 126 is obtained through the $KZ_{(15,5)}$ filter, encompassing both seasonal and long-term components. $X_{ST}(t)$ represents short-term
 127 fluctuations with period less than 33 days in the original time series. $X_{SN}(t)$ is derived as the difference between $X_{BL}(t)$ and
 128 $X_{LT}(t)$, corresponding to seasonal variation on a timescale of months. The KZ filter can fill in missing values by using iterated
 129 moving average technique. Although not all of the ozone measurement sites were active over the entire period 2013–2022,
 130 missing value problems can be handled for most stations after we conduct three iterations with 365-day moving average.

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

135 **2.2.2 Stepwise MLR for separating meteorological influence**

136 As vividly illustrated in **Fig. S1**, a data-based TSM (i.e., MLR integrating the KZ filter) was employed to separate the observed
 137 MDA8 O₃ concentrations into meteorology-driven and emission-driven concentrations (Sadeghi et al., 2022; Shang et al., 2023;
 138 Zhang et al., 2022a). We initially applied the KZ filter to disassemble the MDA8 O₃ time series and all meteorological variables

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

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

143 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
 144 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
 145 term, and ε is the residual term. After establishing MLR models for the short-term and baseline components in each season,
 146 we obtain their respective residual terms. The total residuals, which represent the sum of residuals from baseline variables and
 147 short-term variables, primarily reflect anthropogenic influences. We then applied a $KZ_{(365,3)}$ filter to these aggregated
 148 residuals to derive long-term emission-driven and meteorology-driven O₃ variations. Finally, the meteorology-driven O₃ trends
 149 and emission-driven O₃ trends were obtained through Least Square Method.

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

154 2.2.3 Random forest (RF) for deriving meteorological influence

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

161 RF stands out as a tree-based ensemble learning algorithm adept at handling nonlinear issues and reducing overfitting (Breiman,
 162 2001). An RF model was developed for each state-controlled station in each season to predict the MDA8 O₃ concentration
 163 using the Python package “Sklearn-RandomForestRegressor”. The predictors included six temporal variables (year, month of
 164 a year, day of a week, day of a month, day of a year, Unix time), serving as proxies for anthropogenic emission intensity
 165 (Grange et al., 2018), alongside six MERRA2 meteorological variables as listed in **Table S2** (i.e. SLP, T2max, U10, V10,
 166 RH2, PBLHday). The training dataset comprised 70% of the data, while the remaining 30% was reserved for model evaluation.
 167 A statistical cross-validation technique was employed to determine optimal hyperparameters for enhancing RF prediction
 168 performance (Weng et al., 2022). Coefficient of determination (R^2) values were utilised to assess model performance for each

169 station. Over 70% of state-controlled stations showed $R^2 \geq 0.5$ in all seasons (**Fig. S2b**), which is consistent with the 0.4–0.6
170 range reported in comparable studies (Weng et al., 2022; Lu et al., 2024). Stations with $R^2 < 0.5$ were excluded to avoid
171 significant attribution uncertainty that could be introduced by the RF performance. To evaluate the robustness of the $R^2 \geq 0.5$
172 criterion, we performed sensitivity analyses using thresholds of $R^2 \geq 0.6$ and $R^2 \geq 0.4$, to ensure that our conclusions are not
173 an artifact of an arbitrary cutoff (**Table S3**).

174 After establishing the RF model, both the original time variables and resampled meteorological variables were utilised as input
175 data. For meteorological normalisation, we implemented the protocol of Vu et al. (2019). Meteorological variables were
176 resampled by randomly selecting data from the two weeks before and after the specified date, while temporal proxies remained
177 fixed. To derive the de-weathered MDA8 O₃ concentration for a given day (e.g. March 1, 2013), the random resampling
178 process was iterated 1000 times. The mean predicted O₃ under average meteorological conditions, which refers to de-weathered
179 O₃, corresponds to the emission-driven O₃ concentration. The meteorology-driven MDA8 O₃ concentrations for each season
180 were computed as the difference between observed concentrations and de-weathered concentrations. Detailed processes are
181 shown in **Fig. S2(a)**. The $KZ_{(365,3)}$ filter was then applied to obtain long-term components, and meteorology-driven O₃ trends
182 were derived using Least Square Method.

183 2.2.4 GEOS-Chem (GC) simulation for quantifying meteorological influence

184 The numerical analysis of surface O₃ in China was performed with the GC classic version 13.3.3
185 (<https://github.com/geoschem/GCClassic/releases/tag/13.3.3>). Developed as a global 3-D model, GC incorporates a fully
186 coupled O₃–NO_x–VOCs–aerosol–halogen chemical mechanism, driven by the MERRA2 meteorological input. Numerous
187 studies have leveraged GC to simulate O₃ air quality in China, demonstrating alignment between observational data and model
188 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
189 the long-term surface O₃ concentrations and to quantify the meteorology-driven MDA8 O₃ trends over China. The nested-grid
190 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
191 and 47 vertical layers extending up to an altitude of 0.01 hPa. A global simulation with a horizontal resolution of 2°×2.5°
192 provided the chemical boundary conditions for the nested-grid simulation every 3 hours. To ensure model stability and
193 accuracy, a 6-month spin-up simulation was conducted before the commencement of the targeted 10-year period from March
194 2013 to February 2023.

195 Emissions management within GC is facilitated by the Harmonized Emissions Component, a system introduced by (Lin et al.,
196 2021b). Anthropogenic emissions are sourced from the Community Emissions Data System (CEDS) inventory globally, with
197 specific overwriting by the Multi-resolution Emission Inventory for China (MEIC) within the Chinese region. The simulations
198 for 2021–2022 adopt a similar approach to Zhai et al. (2021), using 2019 MEIC emissions with NO_x emissions reduced by 8
199 ~ 13% and 2017 MEIC with VOCs emissions reduced by 10 ~ 14%, based on the policy released by Ministry of Ecology and

200 Environment of the People's Republic of China. For natural emissions, biogenic VOCs, soil, and lightning NO_x were calculated
201 online in the model. Emissions from biomass burning, ships, and aircraft are sourced from the Global Fire Emissions Database,
202 the CEDS inventory, and the 2019 Aircraft Emissions Inventory Code, respectively.

203 In order to assess the model's performance and to get a quantification of meteorology-driven O₃ trends during the period of
204 2013–2022, two sets of simulations were conducted: (1) BASE: the standard simulation of O₃ concentrations from 2013 to
205 2022, where both meteorological fields and emissions (including anthropogenic, natural, and biomass emissions) vary year by
206 year from 2013 to 2022; (2) FixE2013: a “fixed-emission simulation” where meteorological conditions vary from 2013 to
207 2022 while anthropogenic emissions remain constant at 2013 levels. The FixE2013 simulation is designed to quantify the
208 meteorological influence on O₃ variations. The FixE2013 simulation is designed to obtain the MDA8 O₃ concentrations driven
209 solely by meteorological changes and further quantify the meteorological influence on O₃ variations in four seasons. After
210 applying the KZ_(365,3) filter to derive the long-term meteorology-driven series, trends were calculated through Least Square
211 Method. **Figure S3** evaluates the performance of the GC simulation for 2013–2022. The GC model generally captures the
212 monthly variability in MDA8 O₃ over China and three megacity clusters, with the correlation coefficients greater than 0.80,
213 although it always shows a high bias of surface O₃ in warm seasons (Dai et al., 2024), which can be attributed to its inability
214 to capture the complex terrain, local pollution sources and meteorological conditions, or overestimates of the correlations
215 between the surface O₃ concentration and temperature (Shen et al., 2022; Sun et al., 2021).

216 **2.3 Assessment of uncertainties caused by multi-dataset and multi-method**

217 In this study, the coefficient of variation (CV) is applied to assess the uncertainties in O₃-Meteorology analyses caused by
218 different meteorological datasets or methods. The CV, calculated as the ratio of the standard deviation (SD) to the mean, serves
219 as a statistical metric commonly utilised to measure the diversity within datasets or models (Bedeian and Mossholder, 2000;
220 Chen et al., 2019). Compared to other comparators (e.g. range, inter-quartile range, and SD), the CV is a unit-free measure
221 that quantifies percentage variation relative to the mean and is less sensitive to outliers and heavy-tailed distributions (Högel
222 et al., 1994; Chattamvelli and Shanmugam, 2023). In this study, higher CVs indicate lower consistency of meteorologically
223 driven O₃ trends derived from different datasets or methods. To give a more quantitative assessment, consistency levels were
224 classified as strong and weak with CV<0.5 and CV>1.0, respectively (Wang et al., 2022a). Given the possibility of disparate
225 meteorology-driven O₃ trends detected by different datasets or methods, we consider the absolute value of the CV as a
226 quantitative indicator of the uncertainties. For each season, when examining the uncertainties arising from different datasets,
227 the CV represents the SD of trends derived from the ERA5, MERRA2, and FNL-driven MLR models divided by the mean.
228 Similarly, in the context of multi-method uncertainties, the CV is the SD of trends identified by the MLR, RF, and GC models
229 divided by the mean.

230 **3 Results**

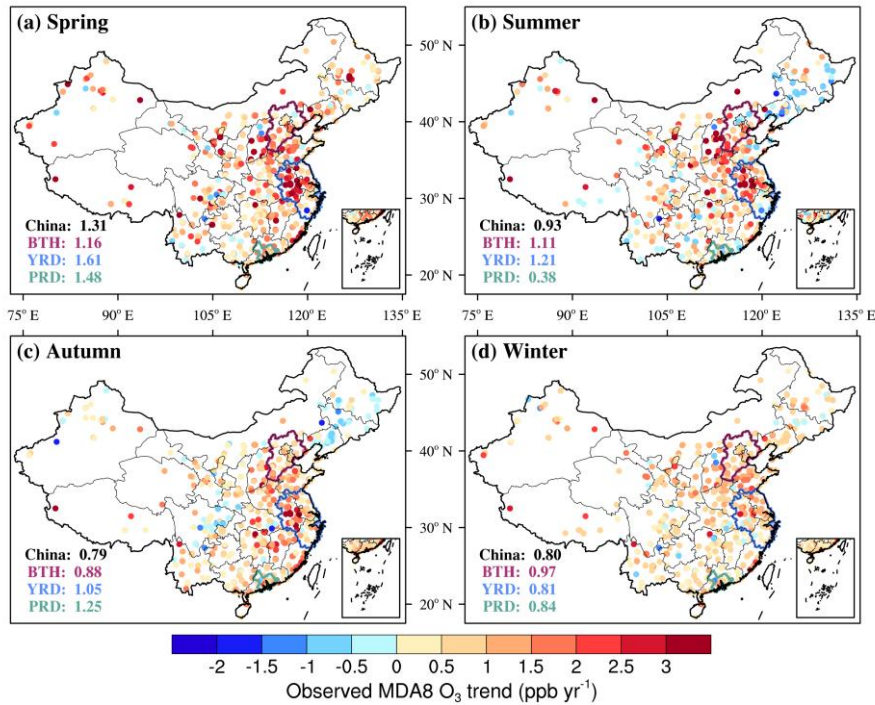
231 **3.1 Observed trends in surface O₃ concentration**

232 **Figure 2** shows the trends in observed MDA8 O₃ concentrations over a 10-year period during four seasons. Noteworthy
233 increases in O₃ concentrations were observed at 78 ~ 93% of state-controlled stations over the years, with the national trend
234 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.

235 The major eastern megacity clusters in China also displayed their highest MDA8 O₃ increase trends in spring, with trends of
236 +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
237 et al., 2024b; Chen et al., 2020; Wang et al., 2022b). During summer, BTH and YRD faced more severe challenges in O₃
238 prevention and control compared to PRD, with rising MDA8 O₃ trends in the former two regions being about three times
239 higher than that in PRD (**Fig. 2b**).

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

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

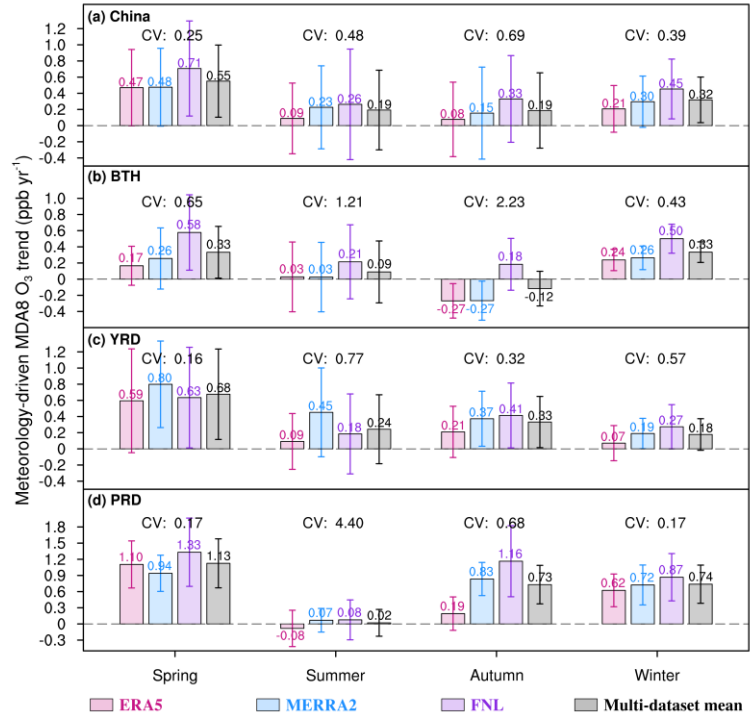


251
 252 **Figure 2.** Trends in observed MDA8 O₃ concentrations in China from 2013 to 2022 during (a) spring, (b) summer, (c) autumn, and (d)
 253 winter. Values in black, purple, blue, and green represent the mean trends for the whole China, BTH, YRD, and PRD, respectively.

254 3.2 Uncertainty in meteorology-driven O₃ trends caused by multi-dataset

255 The traditional statistical method (the MLR model), which has a relatively low computational cost but can provide valuable
 256 insights into the quantification of meteorological contributions to O₃ trends, was used to investigate the uncertainties in O₃-
 257 Meteorology analyses caused by different meteorological datasets. As shown in **Fig. 3(a)**, meteorological conditions contribute
 258 to an increase in MDA8 O₃ concentrations across all seasons in China, with the multi-dataset mean trends ranging from +0.19
 259 (± 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
 260 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
 261 yr⁻¹, and a low CV of 0.25. This suggests a high consistency among the three datasets in assessing the meteorological influence
 262 on surface O₃ concentrations. During summer and autumn, meteorological influences on O₃ show greater spatial heterogeneity
 263 (with higher SD) and larger variability among multi-datasets (with higher CV). Specifically in autumn, the meteorology-driven
 264 O₃ trend derived from the FNL-driven MLR model is 4.1 times larger than that derived from the ERA5-driven MLR model.
 265 Lu et al. (2024) compared meteorology-driven O₃ trends derived from ERA5- and MERRA2-driven MLR models during the
 266 summers of 2013–2019. Their findings revealed that ERA5-derived trends were lower than those from MERRA2 in YRD and
 267 PRD, whereas trends derived from ERA5 were comparable to those from MERRA2 in BTH. This inter-study consensus further
 268 validates the robustness of our methodological framework.

269 **Figure 3(b-d)** depicts the meteorological impact on the MDA8 O₃ trends in the three megacity clusters (BTH, YRD, and PRD).
 270 Meteorology caused the MDA8 O₃ increase in most of the megacity clusters and seasons, except for BTH during autumn. In
 271 seasons where the meteorological effects derived from the three MLR models are all positive, the multi-dataset mean trends
 272 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 +0.73
 273 (± 0.36) to +1.13 (± 0.45) ppb yr⁻¹ in PRD. Consistent with **Fig. 3(a)**, meteorology triggered the most rapid increase in MDA8
 274 O₃ concentrations in spring across the three megacity clusters. The largest meteorological impact in BTH during spring was
 275 also revealed by Luo et al. (2024). Large CVs (>1.0) were observed in BTH during summer and autumn. Notably, the
 276 meteorological influence calculated by the three dataset-driven MLR models even showed opposite trends in BTH during
 277 autumn, indicating challenges in assessing the meteorological impacts on surface O₃ concentrations. In contrast, in YRD and
 278 PRD, the three MLR models demonstrated high consistency across almost all seasons. Although the largest CV reached 4.40
 279 in PRD during summer, it was considered acceptable because the three MLR models indicated that meteorology had a minor
 280 influence (less than +0.1 ppb yr⁻¹) on O₃ trends.



281
 282 **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,
 283 blue, and purple represent trends calculated by ERA5-, MERRA2-, and FNL-driven multiple linear regression (MLR) models, respectively.
 284 The fourth black bar represents the multi-dataset mean trend. Error bars indicate ± 1 standard deviation (SD) of site-level trends calculated
 285 from all available monitoring stations within each region. The absolute value of the coefficient of variation (CV) for each season is also
 286 shown.

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

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

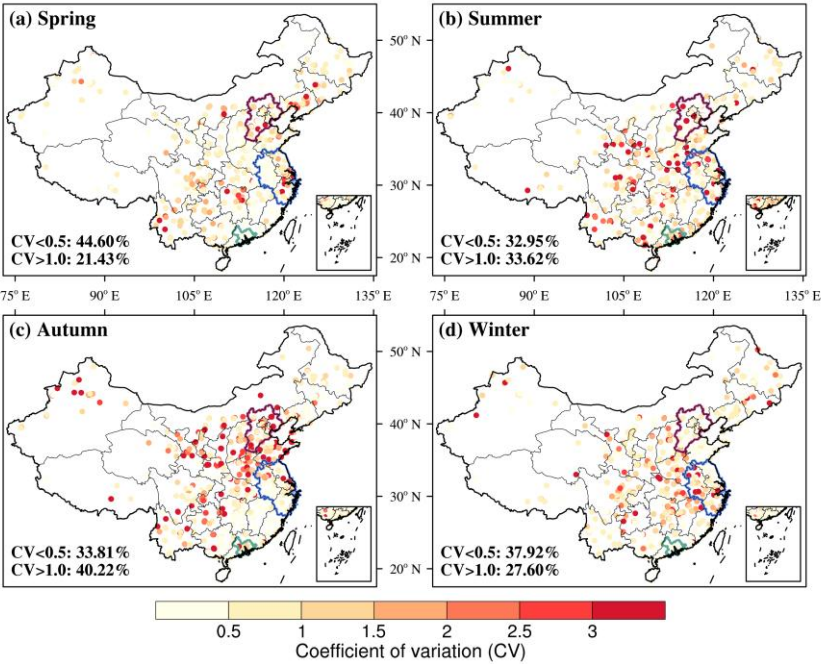


Figure 4. The absolute value of the coefficient of variation (CV) for each state-controlled monitoring station in China during (a) spring, (b) summer, (c) autumn, and (d) winter. The CV is calculated by the standard deviation (SD) of the trends derived from ERA5-, MERRA2-, and FNL-driven MLR models divided 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 shown. The outline marked in purple, blue, and green represents the region of BTH, YRD, and PRD, respectively.

Based on the three dataset-driven MLR models, the meteorological and anthropogenic contributions to the MDA8 O₃ trends in China during 2013–2022 were further examined. As presented in **Fig. 5**, both meteorological conditions and anthropogenic

emissions lead to O₃ increases. According to the ERA5- and MERRA2-driven MLR models, variations in anthropogenic emissions were identified as the dominant factor driving the increase in MDA8 O₃ concentrations across all seasons, with anthropogenic contributions ranging from 63.2% to 90.4%. The results suggest that more stringent emission control policies should be implemented to counteract the adverse effects of meteorological influences on O₃ concentrations.

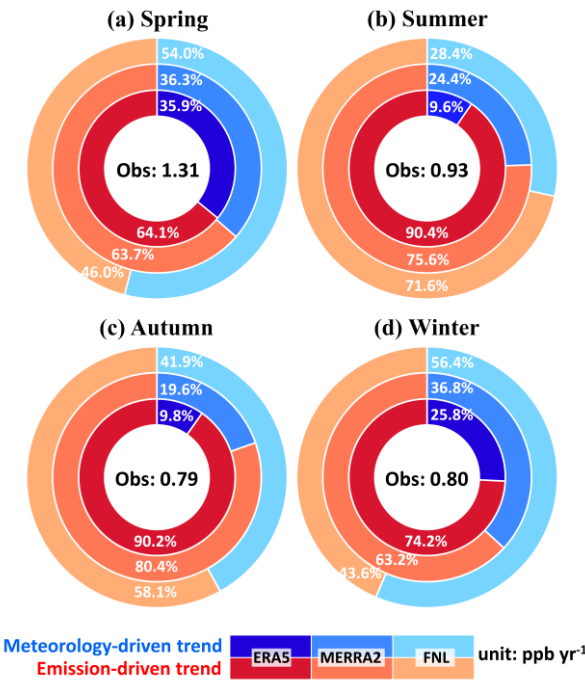


Figure 5. Percentage contributions of meteorological conditions (blue) and anthropogenic emissions (red) to the trends in observed MDA8 O₃ concentrations calculated by ERA5-, MERRA2-, and FNL-driven multiple linear regression (MLR) models in China during (a) spring, (b) summer, (c) autumn, and (d) winter. 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 S4** 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 when separating meteorological and anthropogenic influences on O₃ concentrations in China.

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

This section discusses the uncertainties caused by multi-method (i.e. MLR, RF, GC), all of which are driven by the MERRA2 dataset. **Figure 6** illustrates the meteorology-driven MDA8 O₃ trends calculated by the MLR, RF, and GC models. For the whole of China, the large uncertainties are evident during summer, when the meteorology-driven O₃ trends derived from the MLR model are notably larger than those from the RF and GC models, with a CV of 2.00 (**Fig. 6a**). In the other three seasons, 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 than those computed by the three dataset-driven MLR models (**Fig. 3a**), all models converge on the conclusion that meteorological conditions contribute to the deterioration of O₃ air quality. Meteorology-driven MDA8 O₃ trends exhibited minor variations across different R² thresholds (**Table S3**), indicating that the trends are not an artifact of an arbitrary cutoff.

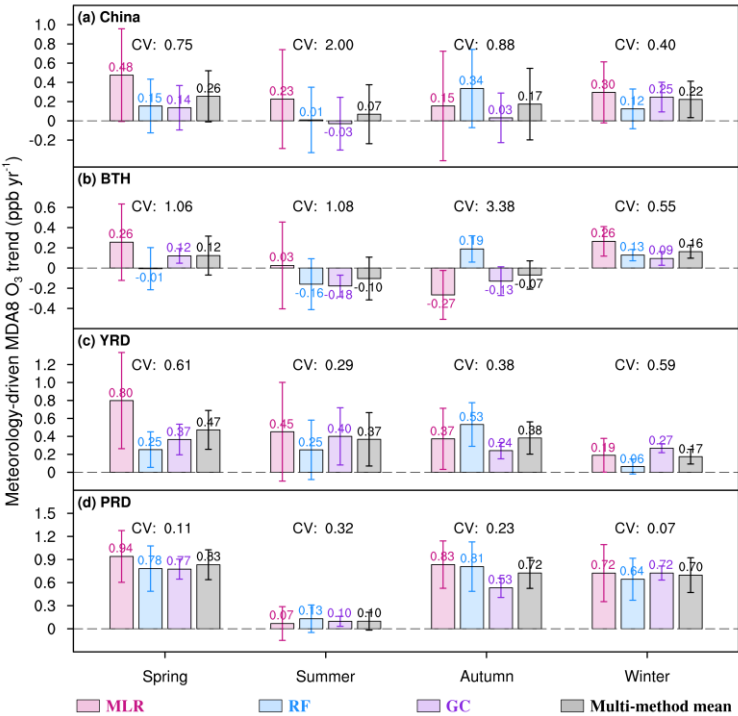
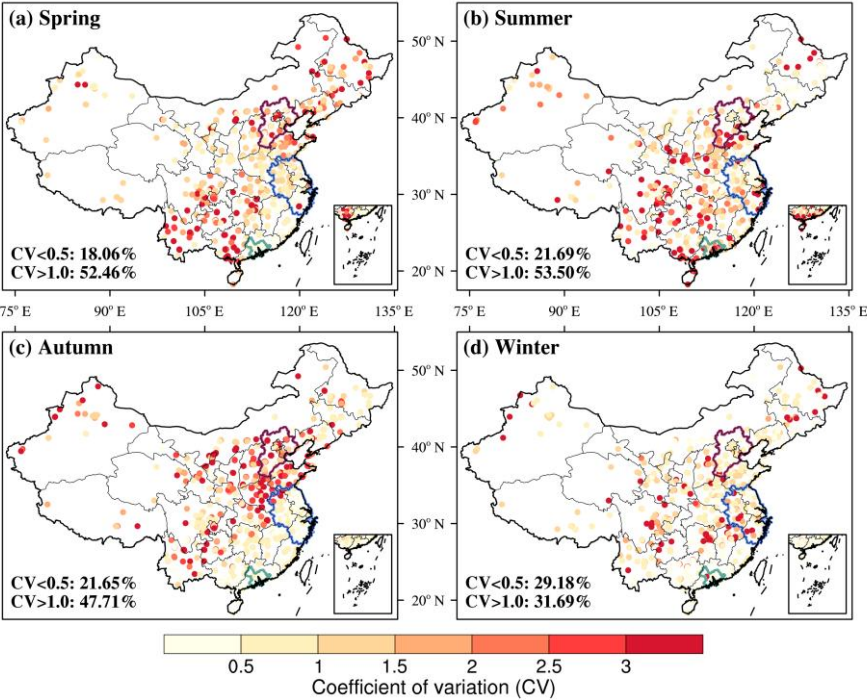


Figure 6. Meteorology-driven MDA8 O₃ trends in (a) the whole China, (b) BTH, (c) YRD, and (d) PRD during four seasons. Values in red, blue, and purple represent trends calculated by multiple linear regression (MLR), random forest (RF), and GEOS-Chem (GC) models, respectively. The fourth black bar represents the multi-method mean trend. Error bars indicate ±1 standard deviation (SD) of site-level trends

341 calculated from all available monitoring stations within each region. The absolute value of the coefficient of variation (CV) for each season
 342 is also shown.

343 In YRD and PRD, the three models exhibit strong agreement across all seasons, with a maximum CV of 0.61. The low
 344 uncertainties are further corroborated by consistent CV estimates derived under different RF's R^2 thresholds (**Table S3**).
 345 Across these regions, where meteorology leads to an increase in O_3 concentrations with multi-method mean trends of $+0.17$
 346 (± 0.08) to $+0.47$ (± 0.22) ppb yr^{-1} in YRD and $+0.10$ (± 0.12) to $+0.83$ (± 0.19) ppb yr^{-1} in PRD. Notably, the most rapid
 347 meteorology-driven O_3 increase is also observed in spring (**Fig. 6c and Fig. 6d**), which is consistent with **Fig. 3c and Fig. 3d**.
 348 Lu et al. (2024) also demonstrated a high degree of consistency among the MLR, ML, and GC models in PRD during summer.
 349 Specifically, all three models indicated that meteorology contributed approximately 25% of O_3 variability over the period
 350 2013–2019. In BTH, the three models perform consistently well only in winter, with meteorology-driven O_3 trends ranging
 351 from $+0.09$ (± 0.07) to $+0.26$ (± 0.15) ppb yr^{-1} and a CV of 0.55. It is also observed that in summer and autumn, meteorology
 352 plays a relatively small role in influencing O_3 air quality despite the controversial results obtained by the three models (**Fig.**
 353 **6b**). This finding aligns with a study focusing on the O_3 air quality in BTH from 2015 to 2022 (Luo et al., 2024), which
 354 suggested that meteorological conditions tend to increase MDA8 O_3 concentration by only $0.01 \mu g m^{-3}$ in summer and decrease
 355 MDA8 O_3 concentration by $0.3 \mu g m^{-3}$ in autumn from 2015 to 2022.



356
 357 **Figure 7.** The absolute value of the coefficient of variation (CV) for each state-controlled monitoring station in China during (a) spring, (b)
 358 summer, (c) autumn, and (d) winter. The CV is calculated by the standard deviation (SD) of the trends derived from multiple linear regression

359 (MLR), random forest (RF), and GEOS-Chem (GC) models divided by the mean. The darker colour means the larger uncertainty in
360 quantifying the meteorological impact on observed O₃ trends. The proportion of state-control stations with CV less than 0.5 and greater than
361 1.0 is also presented. The outline marked in purple, blue, and green represents the region of BTH, YRD, and PRD, respectively.

362 In addition, **Fig. 6** illustrates that the meteorology-driven O₃ trends obtained from GC are relatively smaller. As shown in **Fig.**
363 **S3** and **Table S5**, this difference could partly be attributed to the higher O₃ levels and lower O₃ increases simulated by the GC
364 model before 2018. The GC's systematic overestimation of O₃ concentrations, as well as underestimation of O₃ increases, was
365 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
366 summer of 2000–2019. It is crucial to take into account the overestimation of low-level O₃ observations, as noted in previous
367 studies (Hu et al., 2024c; Mao et al., 2024). To validate this hypothesis, we compared the meteorology-driven O₃ trends
368 calculated by MLR with those calculated by GC from 2018 to 2022, and a higher agreement was found over 2018–2022
369 compared to the 2013–2022 period in **Fig. S9**. The trends driven by RF model are eclectic in more cases (**Fig. 6**) and
370 recommended to isolate meteorological and anthropogenic drivers.

371 From a provincial perspective, as depicted in **Fig. S10**, the three models together indicate that meteorology causes an O₃
372 increase in winter across almost all provinces except for Guizhou and Sichuan. In summer and autumn, meteorology leads to
373 a decrease in 5 provinces, mainly in northeastern China, with trends ranging from -0.42 to -0.11 ppb yr⁻¹. Interestingly, across
374 all seasons, the three models introduce less uncertainty in the developed east coast regions such as Jiangsu, Fujian, and
375 Guangdong compared to other provinces. This suggests that quantifying meteorological impact on O₃ levels in these developed
376 regions along the east coast of China is relatively reliable.

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

382 **4 Limitations**

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

387 Regarding analytical approaches, machine learning efficiently captures nonlinear O₃-meteorology relationships without
388 requiring explicit physicochemical parameterizations, enabling scalable multi-site analysis. However, its inability to resolve

389 chemical mechanisms and sensitivity to predictor selection remain key constraints. Conversely, while GC mechanistically
390 resolves chemistry-transport interactions and enables source attribution, it propagates uncertainties from emission inventories
391 and chemical mechanisms into trend estimates.

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

400 **5 Conclusions and Discussions**

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

407 Over the past decade, increasing trends in MDA8 O₃ were observed at over 78% of state-controlled stations across all seasons,
408 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
409 winter, respectively.

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

419 We further applied the MLR, RF, and GEOS-Chem models to obtain the meteorological influence on O₃ trends to explore the
420 uncertainties caused by multi-method. In China and three megacity clusters, the three methods consistently indicated positive
421 meteorological contributions to O₃ increases during spring and winter, with multi-method mean trends ranging from +0.12 to
422 +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
423 influence was relatively lower, three methods gave conflicting predictions of meteorological influence on O₃ with CVs greater
424 than 1.08. For the whole China, three different methods demonstrated optimal consistency in winter with a CV of 0.40 and the
425 worst consistency in summer with a CV of 2.00. The meteorology-driven O₃ trends obtained from GEOS-Chem model were
426 almost relatively smaller than those obtained by other two methods, which could partly be attributed to the higher O₃ values
427 simulated by the GEOS-Chem model before 2018.

428 All analyses driven by diverse meteorological datasets and analytical methods drew a consistent finding: meteorological
429 conditions almost contribute to O₃ increase across all seasons. The uncertainties of meteorology-driven O₃ trends caused by
430 different analytical methods were larger than those caused by diverse meteorological datasets. Considering that the favourable
431 effects of meteorology on O₃ pollution tend to be weaker after 2019 and the effects of COVID-19, it is necessary to conduct
432 research over different periods and longer periods. In addition, further research is needed to focus on the meteorological
433 contributions to O₃ trends in northern China due to larger uncertainties.

434 **Data availability.** The surface O₃ observations are obtained from <https://quotsoft.net/air/>. The ERA5, MERRA2, FNL, and
435 FNL025 reanalysis meteorological data are taken from <https://cds.climate.copernicus.eu/datasets>,
436 http://geoschemdata.wustl.edu/ExtData/GEOS_0.5x0.625_AS/MERRA-2/, <https://rda.ucar.edu/datasets/d083002/>, and
437 <https://rda.ucar.edu/datasets/ds083-3/>, respectively. The code of the GEOS-Chem (version 13.3.3) model is available at
438 <https://zenodo.org/records/5748260>. The MDA8 O₃ observations and analytical results derived from MLR, RF, and GEOS-
439 Chem can be obtained from <https://doi.org/10.5281/zenodo.15859027>.

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