

Historical and Future changes of surface ozone over China from CMIP6 models, including an assessment of present-day uncertainties in model prediction

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Abstract: Ozone (O₃) contributes to global climate change and poses a direct threat to human health.

This study investigates the historical variability, future projections, and associated present-day uncertainties of surface O₃ concentrations over China using simulations from nine CMIP6 models and observational data from the Tracking Air Pollution in China (TAP) dataset. A multi-model ensemble mean (MME), constructed using an equal-weighted approach, is used to evaluate model uncertainties across different seasons, underlying surface types, total cloud cover, and PM_{2.5} concentrations, and to assess model performance under future Shared Socioeconomic Pathway (SSP) scenarios. The results show that the MME captures the pronounced seasonal cycle of surface O₃, with higher concentrations during June–August (JJA, ~105 μg·m⁻³) and lower values during December–February (DJF, ~55

μg·m⁻³), but underestimates O₃ across most regions of China, particularly in East China. Model performance varies with environmental conditions, showing lower bias, MAE, and RMSE over natural land surfaces than over anthropogenic surfaces. The O₃ bias is minimized under cloudy conditions, maximized under partly cloudy conditions, and generally increases with rising PM_{2.5} concentrations before declining beyond a threshold. Over the historical period, the MME simulates a substantial increase in annual mean surface O₃ across China (~39.3 μg·m⁻³). Future projections indicate continued O₃ increases under weak mitigation (SSP3-7.0), with East China rising by 26.9%, and widespread decreases under strong mitigation (SSP1-2.6), particularly in Southwest and South China (>30 μg·m⁻³). Analysis of model spread and its drivers indicates that uncertainties in surface O₃ projections arise from

the combined effects of emissions (including precursors and PM_{2.5}), climate conditions, and model
35 representations of chemistry and circulation. Improving the understanding of these coupled influences
is essential for enhancing the reliability of regional O₃ projections and for informing effective air
quality and climate mitigation strategies in China.

1 Introduction

40 Ozone (O₃) is one of the most important trace gases components in the Earth's atmosphere, serving as a
protective barrier for the global ecosystem and a crucial heat source in the stratosphere (Zhang et al.,
2017). Variations in ozone concentrations strongly influence the climate of both the stratosphere and
the troposphere (Xie et al., 2017; Haase and Matthes, 2019; Lin and Ming, 2021), and play a critical
role in controlling the atmospheric temperature structure. Studies have shown that 90% of atmospheric
45 O₃ is concentrated in the stratosphere, with only about 10% distributed in the troposphere, however, the
climatic effects caused by tropospheric O₃ variations can be comparable to the perturbations induced in
the stratosphere (Xie and Zhang, 2014). As one of the major sources of OH radicals, O₃ indirectly
determines the lifetime of various trace constituents in the troposphere (Levy, 1971). Additionally, O₃
is an important greenhouse gas, a strong oxidant, and a plant toxin, which not only influences global
50 climate change (Monks et al., 2015) but also directly harms human health (Shindell et al., 2012; Wang
et al., 2021) and vegetation growth (Avnery et al., 2011; Lin et al., 2018; Feng et al., 2018). The Global
Burden of Disease Report (GBDR) states that more than 360,000 premature deaths globally in 2019
were attributed to exposure to ambient O₃, and that high O₃ exposure may exacerbate the
PM_{2.5}-mortality risk (Weichenthal et al., 2017). Therefore, studying the evolution of O₃ is of great
55 significance for understanding global climate change and protecting Earth's ecosystems.

Over the past decade, due to rapid industrial development, precursor pollutants have been continuously
emitted in large quantities, causing severe PM_{2.5} and O₃ pollution in China (Maji et al., 2018; Lu et al.,
2018; Qin et al., 2021). To improve air quality, the State Council issued the "Air Pollution Prevention
and Control Action Plan" (APPCAP) in 2013, with the goal of reducing PM_{2.5} concentrations in the key
60 regions of Beijing-Tianjin-Hebei, the Yangtze River Delta, and the Pearl River Delta by 25%, 20%,
and 15%, respectively, by 2017. Accordingly, a series of air pollution control measures have been
implemented, including optimizing industrial structure, increasing the supply of clean energy, limiting

high-emission vehicles, and banning high-emission sources such as open biomass burning (Qiu et al., 2016). As a result, since the "13th Five-Year Plan", China has made significant progress in air pollution
65 mechanism research and control (Li et al., 2020; Lu et al., 2020; An et al., 2022; Su et al., 2022), particularly with a notable decrease in the annual average concentration of PM_{2.5} in major regions. In stark contrast to the improvements in PM_{2.5} pollution control, most regions in China experienced a persistently fluctuating upward trend in annual O₃ concentrations during 2013–2018, with an average annual increase of 1–3 ppb (Li et al., 2019). By 2018, the national annual mean O₃ concentration had
70 increased by 17.59% and 15.22% compared to 2013 and 2015 levels, respectively (Wang et al., 2020). The proportion of O₃-polluted days has become increasingly significant, and prolonged, large-scale O₃ pollution episodes have occurred more frequently, particularly in major urban agglomerations such as the Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD), and Pearl River Delta (PRD) regions (Dai et al., 2020; Zhao et al., 2020). Notably, since 2015 in the PRD and since 2017 in the YRD, the
75 proportion of days with O₃ exceedance has surpassed that of particulate matter, making O₃ the primary pollutant (Lu et al., 2018; Wang et al., 2020). Despite the positive effects of policy implementation on reducing nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in China (Lu et al., 2018), significant uncertainties remain regarding the abundance, spatial distribution, and related processes of these short-lived gases, which constrain the further optimization and effectiveness of emission control
80 policies (Wild et al., 2020). Against this backdrop, surface O₃ pollution in China continues to worsen and expand from 2015 to 2024 (Wang et al., 2020; Xiao et al., 2022; Figure S8). While PM_{2.5} pollution has shown consistent improvement, effectively controlling O₃ pollution has emerged as a critical challenge for air quality management in China, posing serious difficulties for both the scientific community and policymakers. Under the guidance of China's national "Dual-Carbon" strategy, which
85 targets carbon peaking by 2030 and carbon neutrality by 2060, synergistic control of greenhouse gas emissions and air pollutants has become essential for achieving sustained improvements in air quality. This is because the dominant CO₂ emission sectors (e.g., power generation, industrial production, and transportation) are also major sources of ozone precursors such as NO_x and anthropogenic VOCs. Therefore, reducing fossil fuel combustion can bring co-benefits by mitigating both climate warming
90 and O₃ pollution. Given this context, accurately understanding the spatiotemporal evolution of surface O₃ concentrations in China is of great importance, as it provides a scientific basis for evaluating the effectiveness of emission control measures and for guiding future air quality management strategies.

Currently, ground-based observations (including surface and radiosonde measurements) (Zhan et al., 2021; Liu et al., 2022), satellite remote sensing retrievals (such as column concentrations, vertical profiles, and multi-source data fusion) (Hubert et al., 2021; Zhao et al., 2022), and model simulations (Xue et al., 2020; Morgenstern, 2021) provide essential data and analytical approaches for monitoring O₃ and its precursors, investigating pollution sources and transport characteristics, and evaluating the accuracy of retrieval products. Ground-based observations are known for their high accuracy, satellite remote sensing offers broad coverage, and model simulations can extend both spatially and temporally through parameterization, these methods complement each other and collectively support ozone-related research. For decades, both chemical-climate models (CCMs) and chemical transport models (CTMs) have been indispensable tools for studying global surface O₃. CTMs are widely used for high-resolution regional simulations, while CCMs are particularly suited for investigating long-term global trends, reproducing historical O₃ distributions, predicting future changes across various spatial and temporal scales, and exploring interactions with precursors and atmospheric physical-dynamic processes. However, due to the complexity of the tropospheric O₃ budget mechanisms, particularly the effects of chemical reaction chains, precursor emission distributions, and meteorological conditions, significant discrepancies remain in model results, both among different models and compared with observations. These discrepancies reflect the limitations of the models in parameterization of physical and chemical processes, as well as uncertainties in emission inventories and boundary conditions. Therefore, identifying, investigating, and quantifying the differences between models and observations is crucial for improving and advancing model performance (Young et al., 2018). Beyond individual model studies, international multimodel initiatives have advanced understanding of surface O₃ pollution. The Hemispheric Transport of Air Pollution (HTAP) project (Fiore et al., 2009) and its regional counterpart MICS-Asia underscore the role of hemispheric and transboundary transport in shaping regional O₃ patterns (Li et al., 2019). The Tropospheric O₃ Assessment Report (TOAR) provides a global benchmark for evaluating surface O₃ models and assessing historical trends using harmonized observations and multimodel comparisons (Young et al., 2018). CMIP6 and the latest IPCC AR6 adopt new emission scenarios driven by different socioeconomic pathways, the Shared Socioeconomic Pathways (SSPs), replacing the four Representative Concentration Pathways (RCPs) used in CMIP5, this is a significant advancement in the CMIP6 scenarios (Eyring et al., 2016; Zhou et al., 2019). Previous studies have shown that most CMIP6 Earth

System Models (ESMs) are capable of capturing the spatial distribution of global surface O₃ concentrations (Turnock et al., 2020; Ivanciu et al., 2021; Griffiths et al., 2021; Shang et al., 2021), but they tend to produce an overall positive bias of 5–10% (3.6±4.4 ppbv) (Sun and Archibal, 2021), with larger biases in the Northern Hemisphere and smaller biases in the Southern Hemisphere. This discrepancy may be attributed to the limitations of O₃ precursor emission data (Young et al., 2013). Currently, the evaluation of the latest CMIP6 simulations for surface O₃ in China is still limited. Therefore, this paper based on multi-model O₃ products from CMIP6, conducts an analysis of the historical and future changes of surface O₃ in China, as well as the associated present-day uncertainties. First, using the O₃ dataset from the Tracking Air Pollution in China (TAP), we assess the distribution and uncertainty of surface O₃ simulated by nine CMIP6 models under various conditions, including different temperatures, cloud cover levels, complex land surface types, and pollutant concentrations, for the period 2015–2023 across China and its seven sub-regions (Northeast China, North China, East China, South China, Central China, Northwest China and Southwest China) (Figure 1). Secondly, the changes in surface O₃ over different regions of China during the historical period 1850–2014 are calculated to provide a background for the analysis of future changes. Then, based on different SSPs in CMIP6 experiments, the future changes in surface O₃ across China are predicted and analysed for the period 2015–2100. Finally, a comparison of different CMIP6 models under a single future scenario (SSP3-7.0) is conducted to identify the potential causes of models differences, aiming to provide valuable references for future O₃ pollution control and prediction efforts.

2 Research data and methods

The data used in this paper primarily includes O₃ products simulated by all models (9 models) in the CMIP6 chemistry models ("AERmon" CMIP6 table ID) under both historical and future scenarios (see Table 1 and Table S4). All data can be accessed from the World Climate Research Programme node(WCRP) (<https://esgf-node.llnl.gov/search/cmip6/>, last access: 8 April 2024). Specifically, all available data from 1850 to 2014 were obtained from the historical experiments of CMIP6 (Eyring et al., 2016), which studied the surface O₃ changes in China during the industrial period. Additionally, all available data for the period 2015–2100 from different shared economic pathways in ScenarioMIP (O'Neill et al., 2016), were used, and the specific SSP3-7.0-lowNTCF scenario from AerChemMIP

(Collins et al., 2017) were used to investigate future changes in surface O₃ across different regions of China. To investigate the potential drivers behind the differences in future surface O₃ projections over China and its sub-regions under the SSP3-7.0 scenario, this study further incorporates and analyzes VOCs emission data provided by CMIP6. In CMIP6, variables representing non-methane volatile organic compound (NMVOC) emissions primarily include emivoc and emibvoc. The emivoc variable denotes the total emission rate of NMVOCs, covering both anthropogenic and biogenic sources, whereas emibvoc represents NMVOCs emissions from natural sources (e.g., vegetation), and is commonly used as a proxy for biogenic volatile organic compounds (BVOCs). For clarity and consistency, the terms NMVOCs and BVOCs are hereafter used to refer to the emission fluxes represented by emivoc and emibvoc, respectively.

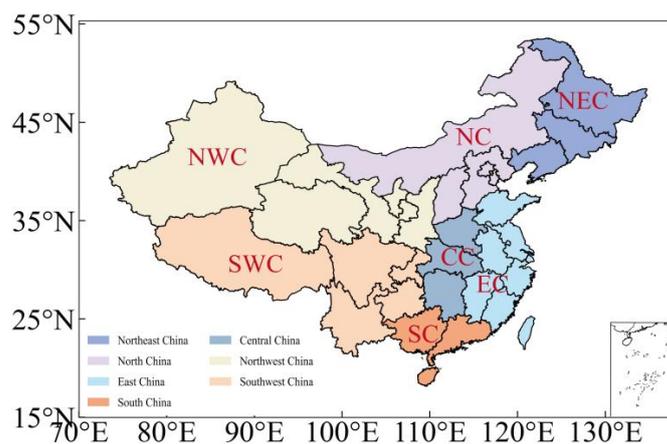
For the historical experiments, CMIP6 provides O₃ data from 9 models and 44 ensemble members. The future scenario with the most available data is SSP3-7.0, with O₃ data from 9 models and 37 ensemble members, followed by SSP3-7.0-lowNTCF, which includes 8 models and 15 ensemble members. For other Tier 1 scenarios (SSP1-2.6 and SSP2-4.5), O₃ data from 5 models are available, while SSP5-8.5 includes 4 models. Due to the limited availability and inconsistent model coverage of Tier 2 CMIP6 scenarios (SSP1-1.9, SSP4-3.4, SSP4-6.0, and SSP5-3.4-OS), all present-day uncertainty analyses and multi-model spread analysis in this study are conducted exclusively using Tier 1 scenarios. Tier 2 scenarios are included in Section 5.1 only to illustrate potential future trajectories of surface O₃ under alternative emission pathways. Some numerical changes are reported to highlight relative differences between these scenarios, but they are not used for robust multi-model inter-scenario comparisons or quantitative uncertainty assessment. Although one CMIP6 model (MRI-ESM2-0) provides simulations for all Tier 1 and Tier 2 scenarios, single-model inter-scenario comparisons are not performed, as our analysis relies on multi-model ensemble statistics to ensure representative and robust conclusions.

To assess the uncertainty in the CMIP6 simulation of present-day O₃ concentrations, this study utilizes the TAP (<http://tapdata.org.cn/>, last access: 8 April 2024) O₃ monthly products for the period from January 2015 to December 2023, with a spatial resolution of 0.1°. TAP is a near-real-time atmospheric composition tracking dataset for China, developed by Tsinghua University in collaboration with several other institutions, which mainly includes O₃, PM_{2.5} and their major chemical components. The O₃ product is derived through a machine learning model that integrates multiple data sources, including observational data, satellite remote sensing vertical profiles of O₃, CMAQ simulations, WRF

simulations, vegetation indices, nighttime lights, and population data, to estimate daily O₃ concentrations. TAP estimates show a high correlation with in-situ observations of maximum daily 8-hour average O₃, with an R² value of up to 0.70 (Xue et al., 2020; Xiao et al., 2022).

185 **Table1. Number of ensemble members used for the historical- and future-scenario experiments from each model in the analysis of surface O₃ in this study.**

CMIP6 Models	Institution	Resolution	Historical	SSP1 -2.6	SSP 2-4.5	SSP3 -7.0	SSP3-7.0-lowNTCF	SSP5 -8.5	SSP1 -1.9	SSP4 -3.4	SSP4 -6.0	SSP5-3.4-OS	Model reference
BCC-ESM1	Beijing Climate Center, China Meteorological Administration, China	2.813° ×2.813°	3			3	3						Wu et al. (2020)
CESM2-WACCM	National Center for Atmospheric Research, Climate and Global Dynamics Laboratory, USA	1.25° ×0.94°	3			3	3						Emmons et al. (2020)
EC-Earth3-AerChem	European Consortium of Meteorological Services, Research Institutes, and High-performance Computing Centers	3.0° ×2.0°	4		1	3	3						Noije et al. (2021)
GFDL-ESM4	NOAA Geophysical Fluid Dynamics Laboratory, USA	1.0° ×1.25°	1	1	1	1	1	1	1				Horowitz et al. (2020)
ISPL-CM5A2-INCA	Institute Pierre Simon Laplace, Paris, France	3.75° ×1.875°	1			1	1						Sepulchre et al. (2020)
MIROC-ES2H	University of Tokyo, National Institute for Environmental Studies, and Japan Agency for Marine - Earth Science and Technology, Japan	2.813° ×2.813°	3	1	2	1		3	1				Hajima et al. (2020)
MRI-ESM2-0	Meteorological Research Institute, Japan	2.813° ×2.813°	10	4	10	5	3	5	5	1	1	1	Yukimoto et al. (2019)
UKESM1-0-LL	Natural Environment Research Council, and Met Office, United Kingdom	1.875° ×1.25°	18	5	5	19	1	4					Sellar et al. (2019)
UKESM1-1-LL	Natural Environment Research Council, and Met Office, United Kingdom	1.875° ×1.25°	1	3		1							Mulcahy et al. (2023)
Total number of models			44	14	19	37	15	13	7	1	1	1	



190 **Figure 1. Geographic locations of China and its seven sub-regions used in this study (Northeast China (NEC), North China (NC), East China (EC), South China (SC), Central China (CC), Northwest China (NWC), and Southwest China (SWC)).**

In this paper, surface O₃ concentration is obtained from the lowest vertical layer of each CMIP6 model. For each model, all available ensemble members are averaged using an equal-weighted approach to
195 obtain the model's simulation result; in the MME calculation, the models are likewise combined using an equal-weighted scheme, which inherently assumes independence among the models. All CMIP6 data are linearly interpolated to a 0.1° horizontal resolution to match the TAP dataset when assessing uncertainties in present-day O₃ simulations. It should be noted that CMIP6 outputs, originally at coarse spatial resolutions (>100 km), retain only large-scale spatial patterns through linear interpolation and
200 do not introduce physically meaningful fine-scale features. As a result, localized concentration peaks, particularly in highly urbanized regions, may be smoothed or underestimated, and the interpolated concentrations are therefore more representative of regional-scale patterns rather than urban-scale variability. This approach has been widely adopted in previous CMIP6-based air quality studies when comparing global model outputs with higher-resolution datasets (Turnock et al., 2020; Su et al., 2022).

205 Internal variability due to ensemble spread was quantified for CMIP6 models with more than two realizations under SSP3-7.0. For each model, the standard deviation (SD) of monthly mean surface O₃ across ensemble members was calculated for each month and then averaged over 2015–2023 to yield a single metric (Table S6). Ensemble means of each model were used for inter-model comparisons. The internal variability (generally 2–4 µg·m⁻³) is smaller than inter-model differences, indicating that it
210 does not compromise the robustness of the multi-model comparison based on ensemble means.

The present-day uncertainty of CMIP6-simulated surface O₃ concentrations is assessed by comparison with TAP observations under different environmental conditions, including underlying surface types, total cloud cover, and PM_{2.5} concentrations and components. To avoid introducing additional uncertainty from the CMIP6 models themselves, independent datasets are used for stratification: Total
215 cloud cover from the ERA5 reanalysis, underlying surface types from the MODIS/MCD12Q1 product (treated as fixed categories), and PM_{2.5} data from the TAP observations (Table S1). To ensure spatial consistency, continuous CMIP6 O₃ fields are linearly interpolated onto the 0.1° TAP grid, while categorical underlying surface types are resampled to the same grid using a nearest-neighbor method to preserve their classification. Therefore, interpolation affects only continuous model fields and does not
220 alter land surface categories, although it may smooth fine-scale gradients in modeled concentrations. To quantify uncertainties, for different underlying surface type, cloud cover category, and PM_{2.5} concentration and its components, the difference between CMIP6 MME and TAP observations is

calculated at each grid cell. The bias, MAE, and RMSE of these differences within each group are used as metrics of model uncertainty.

225 **3 Present-Day O₃ concentrations simulations and uncertainties**

In this section, we first evaluate the inter-model performance and spatiotemporal characteristics of CMIP6 surface O₃ simulations. TAP is then used as an independent reference to assess CMIP6 uncertainties under different environmental conditions, including underlying surface types, cloud cover levels, and PM_{2.5} concentrations and its components (Sections 3.2–3.4).

230 **3.1 Inter-model performance and spatiotemporal evaluation**

Temperature directly influences O₃ production by affecting the rates of photochemical reactions and the emission of plant VOCs, such as isoprene (Coates et al., 2016). Therefore, this study compares and analyzes the multi-year annual mean and seasonal distribution, SD, and biases with TAP data for surface O₃ concentrations over China based on the multi-model Ensemble Mean (MME) of CMIP6 for 235 the period 2015–2023 (Figure 2). It is evident that during summer (June, July, and August; JJA), MME shows higher O₃ concentrations, with a mean value of 105 $\mu\text{g}\cdot\text{m}^{-3}$. This is primarily due to increased photolysis activity, higher levels of oxidants, and enhanced biogenic emissions, all of which promote O₃ formation. Additionally, high temperatures are typically associated with increased atmospheric stability and a reduction in mixing layer height, making it more difficult for O₃ to disperse and dilute, 240 leading to its accumulation near the surface and higher concentrations (Yang et al., 2022). In spring (March, April, and May; MAM) and autumn (September, October, and November; SON), the simulated O₃ concentrations are lower than in JJA, with mean values of 89 $\mu\text{g}\cdot\text{m}^{-3}$ and 78 $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The lowest O₃ concentrations are observed in winter (December, January, and February; DJF), with a mean of 55 $\mu\text{g}\cdot\text{m}^{-3}$.

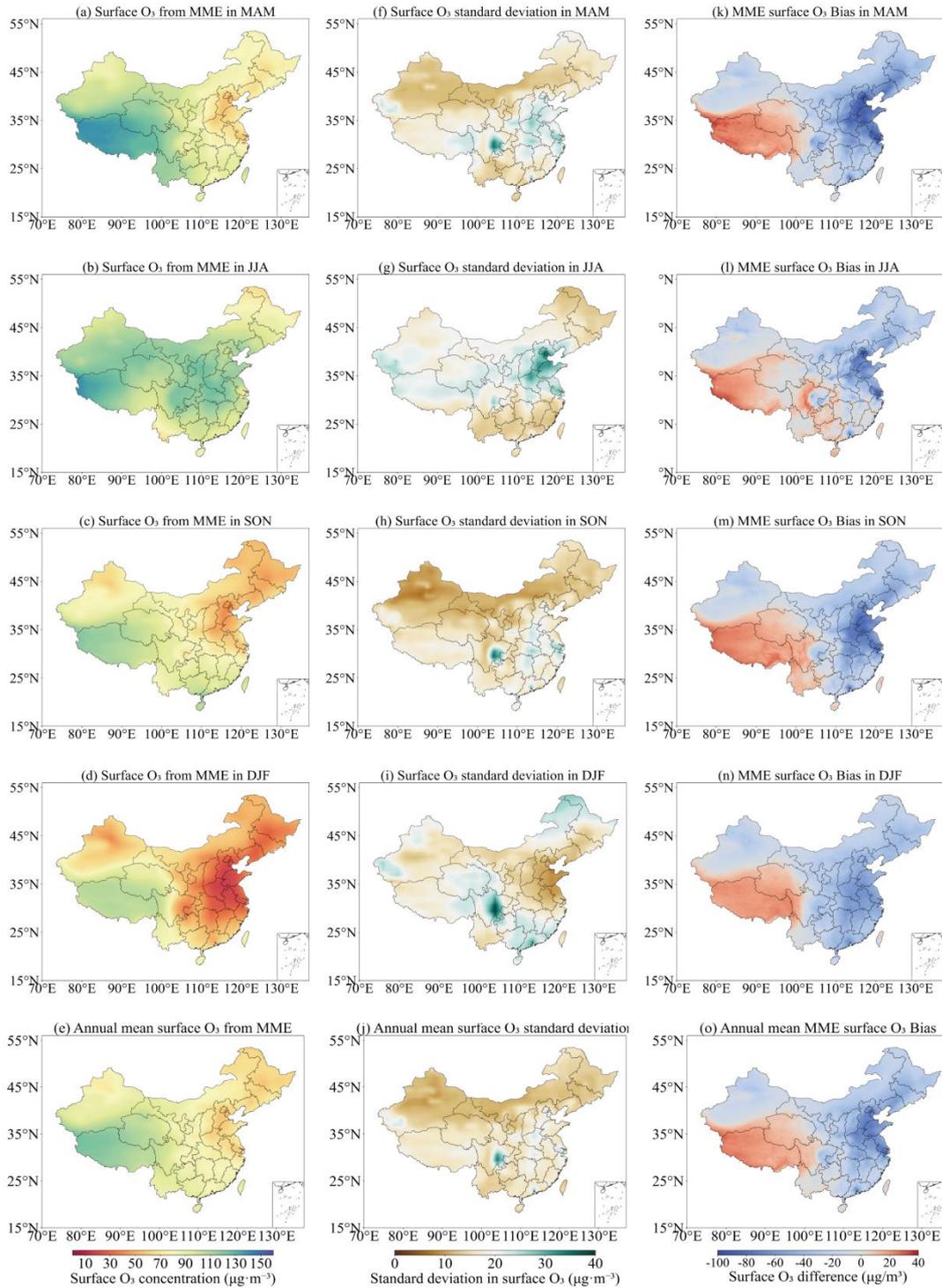
245 For the seven sub-regions (Figure 1), the seasonal patterns are generally consistent with those of the entire China (except for South China). Among them, Central China exhibits the highest O₃ concentration during JJA, reaching up to 117 $\mu\text{g}\cdot\text{m}^{-3}$, while the lowest concentration is simulated in DJF, at only 34 $\mu\text{g}\cdot\text{m}^{-3}$. In South China, O₃ concentrations are higher in SON, with a mean value of 99 $\mu\text{g}\cdot\text{m}^{-3}$, slightly exceeding those in JJA. According to the TAP data, O₃ concentrations in JJA are also 250 lower than those in MAM and SON in South China, and only slightly higher than in DJF. This seasonal

pattern may be related to the southeast monsoon, which has been shown in previous studies to strengthen during JJA and enhance northward transport while increasing precipitation and humidity (Yin et al., 2019), thereby reducing O₃ concentrations. In Southwest China, O₃ concentrations during MAM, SON, and DJF are notably higher than in other regions, particularly in the Tibetan Plateau, where the average O₃ concentration in DJF exceeds 90 µg·m⁻³. This may result from transboundary transport from foreign regions, especially India, where precursor emissions and O₃ concentrations are relatively high (Sahu et al., 2021). In addition, recent modeling work has shown that the Tibetan Plateau receives substantial background O₃ from the free troposphere and long-range transport. Li et al. (2014) demonstrated that trans-Eurasian transport contributes 10–15 ppbv of surface O₃ to western China, superimposed on a natural background of 35–40 ppbv, with particularly strong influence from Indian emissions during the summer monsoon. Furthermore the Tibetan Plateau is also a known hotspot of stratosphere–troposphere exchange (STE), where stratospheric ozone can be transported downward into the troposphere through tropopause folding and subsidence, further contributing to elevated background O₃ levels in this region (Yin et al., 2023). In Northwest China, O₃ concentrations remain high across all four seasons, although a seasonal cycle is present with the highest values in JJA (113 µg·m⁻³). O₃ concentrations in MAM and DJF are the second highest among all regions, following Southwest China. The high O₃ levels can be attributed to multiple factors. First, the region is arid and receives strong solar radiation, which promotes efficient photochemical O₃ production. Second, although overall anthropogenic emissions are relatively low compared with eastern and southern China, some cities such as Urumqi and Yinchuan still exhibit relatively high O₃ concentrations during summer, indicating that local emissions from industrial, energy, and traffic activities cannot be neglected (Zhu et al., 2023). Third, Northwest China can be affected by long-range transport of O₃ and its precursors from surrounding and remote source regions. Modeling studies have shown that European anthropogenic emissions can enhance surface O₃ over northwestern China by approximately 2–6 ppbv during spring and summer, while contributions from the Middle East may occasionally add another 1–4 ppbv (Li et al., 2014). Finally, high-altitude areas are more susceptible to STE, allowing O₃ from the upper troposphere and lower stratosphere to be transported downward, further elevating surface O₃ background levels (Yin et al., 2023). Northeast China, on the other hand, has the lowest O₃ concentrations among all sub-regions, particularly during MAM, JJA, and SON, with concentrations of only 76 µg·m⁻³, 88 µg·m⁻³, and 54 µg·m⁻³, respectively. This can be attributed to several factors: first,

the cooler climate and shorter period of strong solar radiation in northeast China slow down the rate of photochemical ozone production. Second, the region has relatively lower emissions of reactive VOCs compared to the more industrialized and densely populated regions of eastern and southern China, which constrains the overall ozone production potential. Third, northeast China is frequently influenced
285 by clean continental air masses from Siberia, which effectively dilute local and regional pollutant concentrations. In the experimental scenario designed by Zhang et al. (2018), which applied clean-air background concentration boundary conditions and excluded the influence of transboundary anthropogenic emissions from foreign regions, simulated O₃ concentrations decreased significantly over western China but showed little change over eastern China. These results suggest that emissions
290 from external regions primarily affect O₃ levels in western China, whereas domestic emissions remain the dominant contributor to O₃ concentrations in eastern China.

The inter-model SD (Table S2) is largest in DJF (19.0 $\mu\text{g}\cdot\text{m}^{-3}$), but this peak is mainly confined to the Sichuan Basin in Southwest China and parts of South China. In contrast, although the SD in JJA is slightly lower (18.8 $\mu\text{g}\cdot\text{m}^{-3}$), elevated values extend across a much broader area, particularly in North
295 China and East China. The SD decreases in MAM (16.8 $\mu\text{g}\cdot\text{m}^{-3}$) and is smallest in SON (15.2 $\mu\text{g}\cdot\text{m}^{-3}$). This suggests greater diversity in the seasonal O₃ cycles simulated by individual models during DJF, especially for UKESM1-0-LL and UKESM1-1-LL, which exhibit the most distinct seasonal cycles among the nine models (Figure 3), with significant negative biases in simulated O₃ concentrations during DJF.

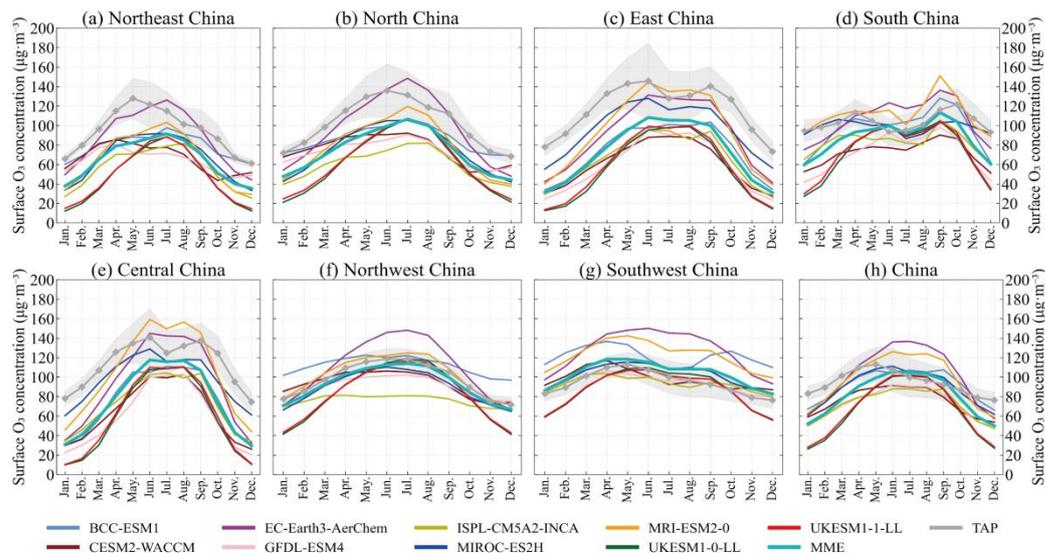
300 The spatial distribution of the MME bias reveals distinct seasonal and regional patterns. On a national average, MME underestimate surface O₃ concentrations, with the largest negative biases occurring in MAM (-25.4 $\mu\text{g}\cdot\text{m}^{-3}$) and DJF(-25.3 $\mu\text{g}\cdot\text{m}^{-3}$), and smaller ones in SON(-22.9 $\mu\text{g}\cdot\text{m}^{-3}$) and JJA (-12.8 $\mu\text{g}\cdot\text{m}^{-3}$). Spatially, the most pronounced underestimations are found over eastern and northern China, particularly during MAM and SON. In contrast, positive biases (overestimations) are evident in
305 Southwest China and in parts of Northwest China (primarily Qinghai Province), especially during SON. Notably, Northwest China (excluding the Qinghai overestimation) exhibits the smallest absolute biases among all sub-regions, with simulated O₃ concentrations aligning most closely with the TAP data across all seasons.



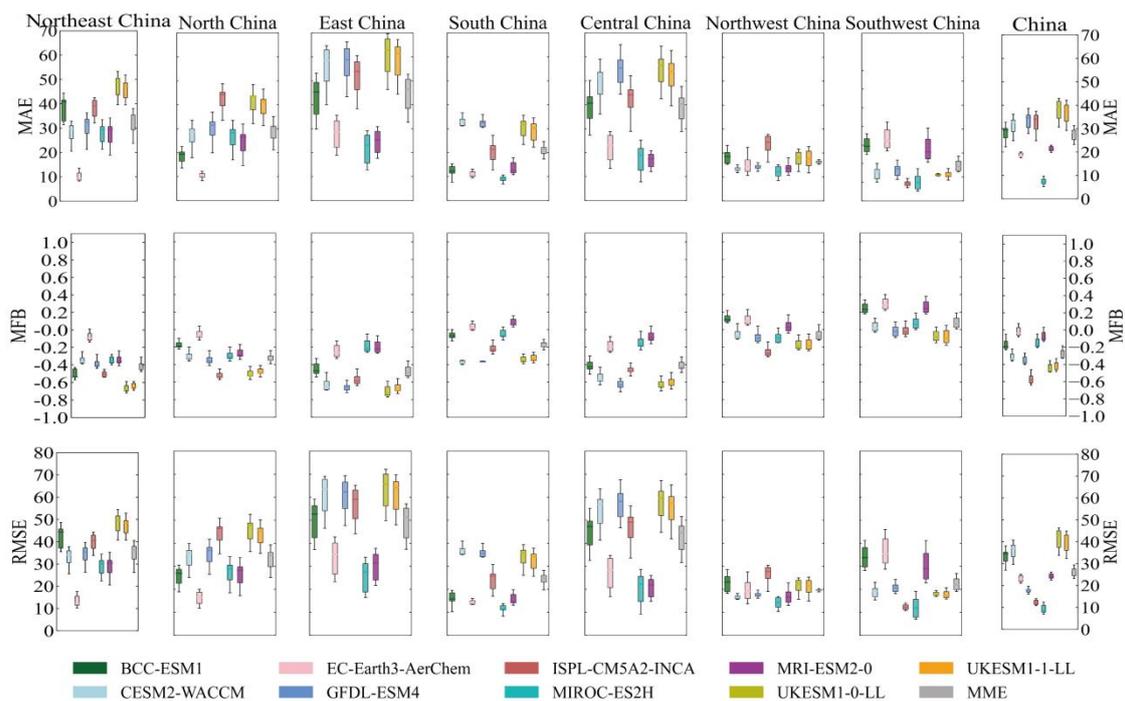
310 **Figure 2. Multi-model (nine CMIP6 models) annual and seasonal mean surface O₃ concentrations over the 2015–2023 period in (a) MAM; (b) JJA; (c) SON; (d) DJF; and (e) annual mean. The SD of the MME in (f)MAM, (g) DJF, (h) JJA, (i)SON, and (j) annual mean. The difference between the MME and TAP observations in (k)MAM, (l) DJF, (m) JJA, (n)SON, and (o) annual mean.**

315 The annual cycles of surface O₃ concentrations in China and its sub-regions simulated by the nine CMIP6 models were compared with the TAP-derived values (Figure 3), it can be seen that the

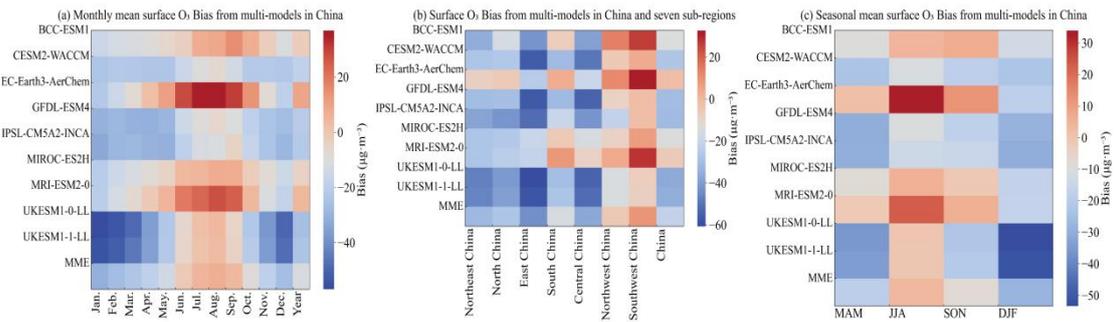
correlation between the two is generally good in most regions ($r > 0.73$), which suggests that the seasonality of the circulation patterns, stratosphere-troposphere exchange, and natural emissions are well captured (Table S3). However, (1) The timing of O₃ peak concentrations in the MME (mostly in July–August, except for East and Central China in June, Southwest China in April) is overall slightly delayed compared to TAP (mostly in May–June), which is consistent with the results of the ACCMIP models (Young et al., 2018). (2) The nine CMIP6 models evaluated in this study generally underestimate O₃ concentrations across most sub-regions of China. The most pronounced underestimations occur in UKESM1-0-LL and UKESM1-1-LL. Although these two models show slight overestimations in Southwest China from June to September, as well as in South China in June and in Northwest China in August, underestimation remains the dominant pattern, particularly in East and Central China during MAM and DJF, where simulated O₃ concentrations are over 60 $\mu\text{g}\cdot\text{m}^{-3}$ lower than the TAP values. This is consistent with the findings of Turnock et al. (2020), who suggested that these biases may be related to excessive NO_x titration in the UKESM1-0-LL model, leading to an underestimation of surface O₃ concentrations over much of the Northern Hemisphere continental regions during DJF. Liu et al. (2022) further conducted a global evaluation of surface O₃ biases in UKESM1, identifying temperature, photolysis rates, and regional chemical composition as key predictors, which may help explain the underestimations identified in this study. (3) In contrast, a small number of CMIP6 models evaluated in this study, including BCC-ESM1, EC-Earth3-AerChem, and MRI-ESM2-0, show a certain degree of overestimation in surface O₃ concentrations throughout the year in Southwest China (with an average overestimation of 30 $\mu\text{g}\cdot\text{m}^{-3}$), Northwest China (with an average overestimation of 10 $\mu\text{g}\cdot\text{m}^{-3}$), and during JJA in South China. This may result from common sources of error in the models, such as uncertainties in emission inventories, deposition processes, or vertical mixing (Wild et al., 2020). Additionally, the coarse resolution of ESMs may lead to overestimation of O₃ concentrations in polluted areas, while higher-resolution models and better consistency between nested models may improve the accuracy of simulated surface O₃ concentrations (Neal et al., 2017).



345 **Figure 3. Comparison of the annual cycle of O₃ concentrations, between individual CMIP6 models, the MME and TAP in China and sub-regions for the period 2015–2023. The grey shading shows SD of TAP observations within the region.**



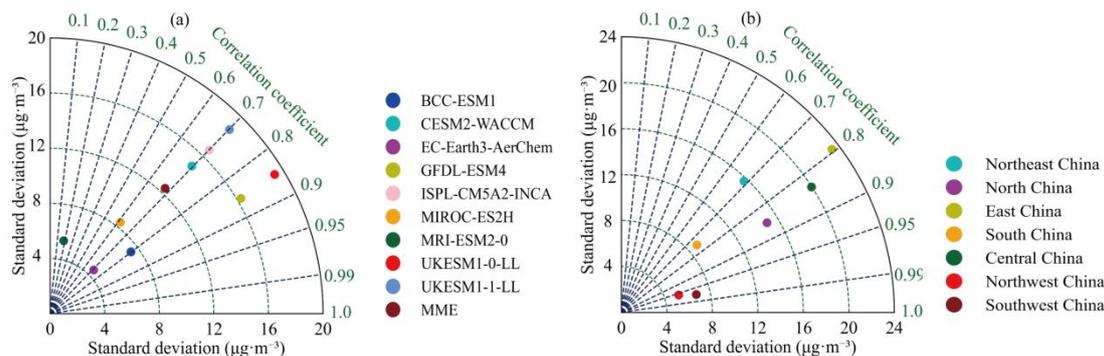
350 **Figure 4. Distribution of differences for O₃ concentrations ($\mu\text{g}\cdot\text{m}^{-3}$) from nine CMIP6 models in China and sub-regions during 2015–2023. The box plots show the 25th and 75th percentiles as solid boxes, median values as solid lines, dots represent the concentrations from MAE, MFB and RMSE, and whiskers extending to the minimum and maximum.**



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Figure 5. Heatmap of O₃ concentrations bias from nine CMIP6 models compared to TAP across different months, seasons and regions in China and sub-regions.

Figure 4 presents a comparison of the Mean Absolute Error (MAE), Mean Fractional bias (MFB), and
 360 Root Mean Square Error (RMSE) between the CMIP6 models and TAP for surface O₃ concentrations. Combined with the bias for different months and sub-regions (Figure 5), correlation coefficients, and SD (Figure 6), it can be observed that the MME simulates the O₃ concentration for China with the smallest bias in June and the largest in January. Among the sub-regions, the simulation results for Northwest China are the most accurate, with high correlation, and the smallest MAE, MFB, RMSE, SD,
 365 and bias. In contrast, the largest MAE, MFB, RMSE, SD, and bias are found in East China, particularly in the autumn and winter. For individual models, EC-Earth3-AerChem shows the smallest annual average bias for O₃ concentrations in China, with an MFB close to zero; BCC-ESM1 exhibits the best correlation; MIROC-ES2H has the smallest MAE and RMSE, providing relatively good simulation results; while UKESM1-0-LL has the largest MAE, MFB, RMSE, SD, and bias. Among the
 370 sub-regions, MIROC-ES2H provides relatively good simulations of O₃ concentrations in South China, while UKESM1-0-LL shows the largest MAE, MFB, RMSE, SD, and bias in East China. Thus, Overall, the MME of CMIP6 performs better in simulating O₃ concentrations during JJA, with larger discrepancies observed in DJF. The simulation in Northwest China is closest to TAP, while the largest discrepancies occur in East China. EC-Earth3-AerChem is better suited for simulating or forecasting
 375 the annual average O₃ concentrations over China, while MIROC-ES2H is more appropriate for error-sensitive scenario analyses, BCC-ESM1 demonstrates superior performance in terms of correlation and temporal consistency, whereas both UKESM1-0-LL and UKESM1-1-LL exhibit higher simulation uncertainties.



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Figure 6. Taylor diagram of the annual mean surface O₃ concentrations simulated by nine CMIP6 models compared with the TAP data during 2015–2023 in China and sub-regions. The radial coordinate shows the SD in the spatial pattern, normalized by the observed SD. The azimuthal variable shows the correlation of the modeled spatial pattern with the observed spatial pattern.

385

3.2 Under different underlying surface types

Vegetation type, land cover, and land use changes can influence biogenic emissions, which in turn affect the accuracy of model-simulated O₃ concentrations (Ashworth et al., 2012). To investigate the impact of these factors on simulation discrepancies and minimize the interference of temperature changes, this study focuses on the JJA period in Northwest China, which has similar climatic conditions and complex surface types. The selected typical underlying surfaces include natural land surfaces such as grassland, forest, desert, and snow and ice (perennial snow), as well as anthropogenic land surfaces such as cropland and urban. By comparing the bias, MAE, RMSE, and SD of MME-simulated surface O₃ concentrations relative to TAP (Figure 7), it is observed that TAP simulates lower O₃ concentrations over natural land surfaces compared to anthropogenic land surfaces, and the MME simulations generally follow this trend. However, the MME simulation results show the highest O₃ concentrations over snow and ice surfaces and the lowest over cropland surfaces. Overall, the MME simulations exhibit lower bias, MAE, and RMSE for natural land surfaces compared to anthropogenic land surfaces, with the best performance over forest and desert surfaces, and the worst performance over urban surfaces, followed by cropland and snow and ice surfaces.

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Natural land surfaces tend to provide more consistent physical and chemical conditions, with limited direct human interference, leading to relatively more accurate simulations of surface O₃ concentrations. Although natural ecosystems can involve complex processes, such as BVOCs emissions from vegetation, deforestation, or wildfires, our analysis based on long-term (JJA) MME effectively smooths

405 out the random uncertainties associated with such transient events. Consequently, the underlying stability of natural surfaces in our study region becomes the dominant feature. In Northwest China during JJA the dominant natural land types (grassland, desert, forest, and snow and ice) generally undergo limited land-use change and exhibit weaker anthropogenic disturbances compared with cropland and urban surfaces. Under these relatively stable conditions, biogenic emission patterns are more predictable, as demonstrated by global modeling studies using MEGAN2.1 (Guenther et al., 410 2012). In addition, dry deposition, a major sink of tropospheric O₃, is more reliably captured over natural surfaces, where vegetation cover and leaf area index are relatively stable. Accurate parameterization of both stomatal and non-stomatal deposition pathways ensures reliable O₃ removal, reducing bias, MAE, and RMSE in multi-model ensemble simulations (Val Martin et al., 2014). In contrast, urban surfaces, characterized by intensive human activities and diverse anthropogenic 415 emission sources (e.g., transportation and industry), present a more complex environment for O₃ formation and destruction. The heterogeneous land cover complicates dry deposition parameterization, often leading to underestimation of O₃ removal and larger discrepancies in simulated concentrations. Snow and ice surfaces can influence O₃ dry deposition during the cold season, particularly in northern 420 China where snow cover often persists for several months. Their high albedo reduces the absorption of solar radiation at the surface and weakens photochemical activity, including the photolysis of NO₂ that contributes to O₃ formation. In addition, snow and ice generally exhibit lower O₃ uptake capacity, with deposition velocities (0.03 cm·s⁻¹) typically smaller than those over vegetated or soil surfaces (Wesely et al., 1981). The magnitude and direction of O₃ flux over snow also depend on factors such as 425 deposited trace gases, solar irradiance, snow temperature, and the underlying substrate (Helmig et al., 2007), but current atmospheric chemistry models often lack detailed parameterizations for these processes. As a result, O₃ deposition over snow and ice remains a potential source of uncertainty for cold-season simulations. It should also be noted that, in the context of O₃ removal, wet deposition contributes minimally to O₃ removal because of its low solubility. Therefore, while dry deposition over 430 snow and ice may be locally important in winter or high-altitude regions, wet deposition has little impact on surface O₃ concentrations.

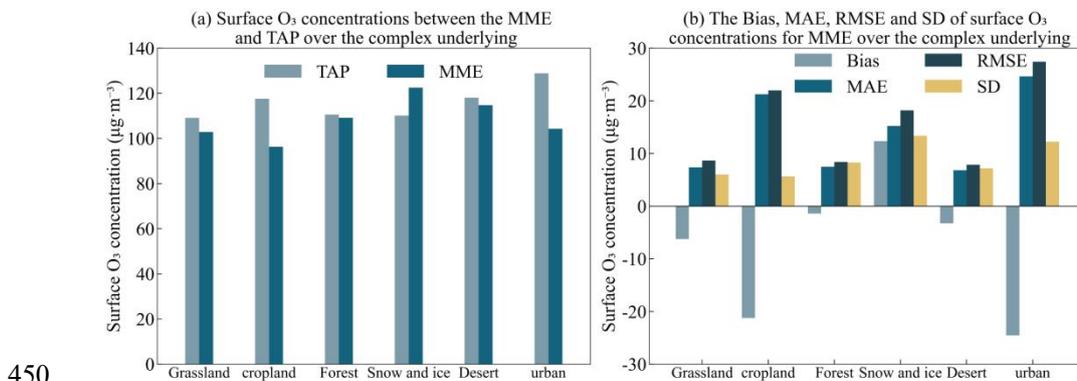
The simulation bias over cropland may stem from the fact that croplands are often associated with agricultural activities (e.g., fertilization and irrigation), which release large amounts of NO_x and VOCs, thereby increasing the complexity of O₃ formation. Furthermore, changes in vegetation types and

435 management practices in agricultural land can also influence biogenic emissions, further affecting the simulation of O_3 concentrations and leading to larger model biases.

The simulation biases identified over different surfaces highlight potential challenges for modeling surface O_3 in China. Specifically, the high errors over cropland and urban areas likely reflect the need for improved model representation of regionally characteristic anthropogenic activities and

440 heterogeneous land cover. Meanwhile, uncertainties over snow and ice surfaces point to gaps in modeling cold-season processes, critical for high-altitude and northern parts of China. While global models may not resolve all local processes, analyzing biases by land cover type provides crucial insight into how these regionally characteristic processes, such as the interplay of anthropogenic and biogenic emissions, deposition, and meteorology, govern surface O_3 patterns. This regional perspective helps

445 interpret model-observation discrepancies in China and provides context for understanding O_3 simulation uncertainties related to land cover. These findings suggest that enhancing parameterizations for characteristic land-surface processes could improve the reliability of global and regional models applied.



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Figure 7. The biases, MAE, RMSE and SD of surface O_3 concentrations simulated by CMIP6 models relative to TAP from 2015 to 2023 over the complex underlying surface in China.

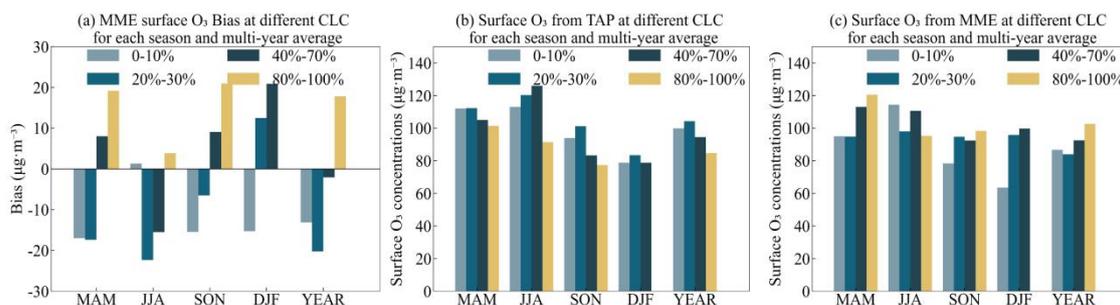
3.3 Under different level of total cloud cover

455 The analysis of surface O_3 bias between MME and TAP under different cloud conditions (Fig. 8) is based on total cloud cover from the ERA5 monthly reanalysis dataset. Using these data, and following the classification of Han and Cong (2015), conditions were categorized into four levels: clear sky (0–10%), partly cloudy (20–30%), cloudy (40–70%), and overcast (80–100%). It is important to note

that while ERA5 provides observation-constrained, physically consistent fields suitable for this
460 diagnostic purpose, uncertainties inherent to any reanalysis (e.g., from retrieval errors and model
assumptions) remain. Nonetheless, ERA5 is considered one of the most reliable datasets for studying
cloud-meteorology interactions (Wu et al., 2023), supporting the robustness of our comparative
analysis. Analysis results show that the TAP simulation exhibits the highest surface O₃ concentrations
under partly cloudy conditions, while under other cloud cover categories, O₃ concentrations generally
465 decrease with increasing total cloud cover (except during JJA). This pattern is primarily attributed to
the attenuation of solar radiation by clouds, which suppresses photochemical O₃ production. In
addition, accompanying precipitation processes can influence atmospheric stability and boundary layer
development, which may indirectly affect surface O₃ concentrations. Under partly cloudy conditions,
however, the atmosphere is generally more stable with weaker vertical mixing, allowing O₃ to
470 accumulate near the surface. During JJA, the relationship becomes more complex, high pollutant loads
and intricate meteorological conditions likely counteract the influence of total cloud cover, leading to a
less straightforward association between cloud amount and O₃ concentrations.

In contrast, the MME simulations do not fully reproduce this pattern. The annual mean bias is smallest
under cloudy conditions and largest under partly cloudy conditions. On a seasonal scale, the smallest
475 bias occurs under clear-sky conditions during JJA, while the largest bias is found under partly cloudy
conditions. These discrepancies may stem from the complex interactions through which cloud cover
modulates shortwave radiation, photochemical rates, and meteorological variables such as temperature,
precipitation, and boundary layer height. They are also closely tied to structural differences among
models in physical parameterizations, radiative transfer schemes, and chemical mechanisms.

480 Therefore, when using CMIP6 models for O₃-related assessment and projection, it is important to
consider how cloud cover modulates shortwave radiation and photochemical reaction rates, how
precipitation affects atmospheric stability and boundary layer development, and how other
meteorological variables such as temperature, humidity, and wind influence both chemical reactions
and pollutant transport. Proper attention to these interactions is particularly crucial under polluted and
485 complex meteorological conditions, where the parameterization of these coupled processes in current
models remains challenging. Improving their representation is therefore key to reducing systematic
uncertainties and enhancing projection accuracy (Jacob and Winner, 2009).



490 **Figure 8. The bias in surface O₃ concentrations simulated by CMIP6 models relative to TAP from 2015 to 2023 under different total cloud cover levels in China.**

3.4 Under different concentrations of PM_{2.5} and its components

Aerosols play a crucial role in the simulation of surface O₃ concentrations. Variations in PM_{2.5} can influence O₃ formation through several pathways: (1) chemical effects: changes in aerosol composition (e.g., sulfate, nitrate, organic matter, BC) can perturb the concentrations of key radicals (e.g., OH, HO₂) and thus alter photochemical reaction rates (Wang et al., 2019); (2) radiative effects: aerosols modify optical properties and reduce actinic flux at the surface, thereby suppressing photolysis rates and slowing O₃ production; and (3) heterogeneous effects: reactions on aerosol surfaces can directly consume NO_x or other precursors, shifting the chemical environment for O₃ formation (Qu et al., 2021).

500 Lou et al. (2014) showed that considering aerosols reduces the mean bias in simulated O₃ over China from 33% to 9% using GEOS-Chem. Therefore, this study analyzes the bias in surface O₃ concentrations simulated by the MME relative to TAP under different PM_{2.5} levels (Figure 9). The results show that during the JJA and SON, when PM_{2.5} concentrations are relatively low, TAP data indicate an increase in O₃ concentrations with rising PM_{2.5} levels. However, during the DJF, when PM_{2.5} concentrations are higher, O₃ concentrations decrease as PM_{2.5} levels increase. This is primarily due to the fact that, during JJA and SON, although the increases in PM_{2.5} concentrations can attenuate solar radiation and participate in heterogeneous uptake of HO₂ and RO₂ radicals. However, their overall inhibitory effect on O₃ formation is relatively weaker during these seasons, because abundant sunlight and strong photochemical activity compensate for aerosol-induced reductions in photolysis rates. In addition, O₃ formation is typically in a VOC-limited regime, under which heterogeneous radical loss plays a smaller relative role (Liu et al., 2019). In contrast, during DJF, limited solar radiation, stronger atmospheric stability, and elevated NO_x concentrations amplify the aerosol impacts, reduced photolysis rates become more critical, heterogeneous radical uptake more effectively suppresses the HO_x cycle,

510

and NO_x titration is more likely to occur. Meanwhile, the increase in BC concentration in PM_{2.5} (Figure 515 S1) enhances the light absorption of PM_{2.5}, further reducing UV radiation and inhibiting O₃ photochemical production. Additionally, other components of PM_{2.5}, such as NO₃⁻ and OM, may influence O₃ through specific pathways, particulate nitrate photolysis generates NO₂ and OH radicals, promoting O₃ formation, while OM and organic nitrates modulate radical (RO_x/NO_x) cycling and NO_x partitioning, thereby affecting O₃ photochemistry (Li et al., 2025). Observational evidence from Chen 520 et al. (2021) in Delhi supports this seasonal behavior: reductions in PM_{2.5} significantly enhance solar radiation, reduce AOD, and increase the photolysis rate of key O₃ precursors, thereby exacerbating O₃ accumulation under VOC-limited conditions, particularly in winter, when AOD is reduced by 50%, O₃ generation can increase by 25%.

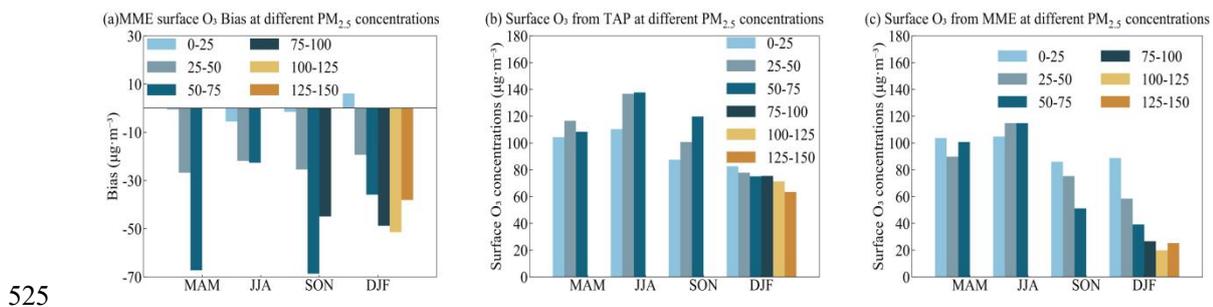


Figure 9. The bias in surface O₃ concentrations simulated by CMIP6 models relative to TAP from 2015 to 2023, under different PM_{2.5} concentrations in China.

The MME simulation results indicate that surface O₃ concentrations generally decrease with increasing 530 PM_{2.5} levels (except during JJA). However, under extreme pollution conditions in DJF, O₃ concentrations exhibit a slight increase once PM_{2.5} exceeds 125 μg·m⁻³. This suggests a complex nonlinear relationship between PM_{2.5} and O₃ formation. Previous studies have shown that reducing aerosol emissions without corresponding cuts in precursor pollutants could lead to increased surface O₃ over eastern China (Li et al., 2018), indicating that aerosols suppress O₃ production through light 535 attenuation and heterogeneous reactions. At low to moderate PM_{2.5} concentrations, the increase in PM_{2.5} largely inhibits O₃ formation by scavenging key radicals (e.g., HO₂ and NO_x) and reducing solar radiation intensity, thereby leading to a decline in O₃. However, under extreme pollution conditions, especially in winter, this suppressing effect tends to saturate. Meanwhile, the nocturnal titration of O₃ is weakened under high NO_x conditions, which, combined with unfavorable meteorological conditions

540 such as temperature inversions and a lower boundary layer, results in O₃ concentrations no longer decreasing with further increases in PM_{2.5} and may even slightly increase.

These findings highlight that the synergistic and inhibitory effects between PM_{2.5} and O₃ vary significantly across seasons and pollution levels. This implies that future air pollution control strategies should adopt coordinated mitigation of both PM_{2.5} and O₃ precursors to avoid potential side effects
545 from single-pollutant reduction policies.

4 O₃ from the pre-industrial period to present day

This study analyzes the annual mean surface O₃ concentration changes in China and its sub-regions from pre-industrial to present times based on 9 CMIP6 models and MME relative to the 2015–2023 mean (Figure 10). The MME results show that, since 1850, the annual mean surface O₃ concentration
550 in China has increased by $39.3 \pm 14.4 \mu\text{g}\cdot\text{m}^{-3}$ (± 1 SD), with the maximum change of $57.9 \mu\text{g}\cdot\text{m}^{-3}$ (from the MIROC-ES2H model) and the minimum change of $23.1 \mu\text{g}\cdot\text{m}^{-3}$ (from the UKESM1-0-LL model). Before 1950, the annual mean increase in O₃ concentration was relatively slow, at only $0.12 \mu\text{g}\cdot\text{m}^{-3}$; however, after 1950, the rate of increase accelerated significantly, with an annual mean increase of $0.28 \mu\text{g}\cdot\text{m}^{-3}$. This change is likely primarily related to the significant increase in anthropogenic activities
555 during this period, especially the substantial increase in anthropogenic precursor emissions, such as CH₄, NO_x, CO, and NMVOCs. The simulations of historical O₃ concentrations by different CMIP6 models show that the EC-Earth3-AerChem model yields the highest values, while the IPSL-CM5A2-INCA model yields the lowest. These differences reflect variations in aerosol, climate, and atmospheric chemistry process simulations across different models. Griffiths et al. (2021), based
560 on ground, sounding, and satellite data from the past few decades, assess the performance of multiple CMIP6 models in simulating surface O₃ concentrations. Their study indicates that these models are capable of accurately reproducing the spatial distribution, seasonal variation, and interannual variability and trends of surface O₃ concentrations. This also indicates the reliability of CMIP6 models in simulating historical surface O₃ variations, which provides support for their future projections of O₃
565 concentrations under climate change scenarios.

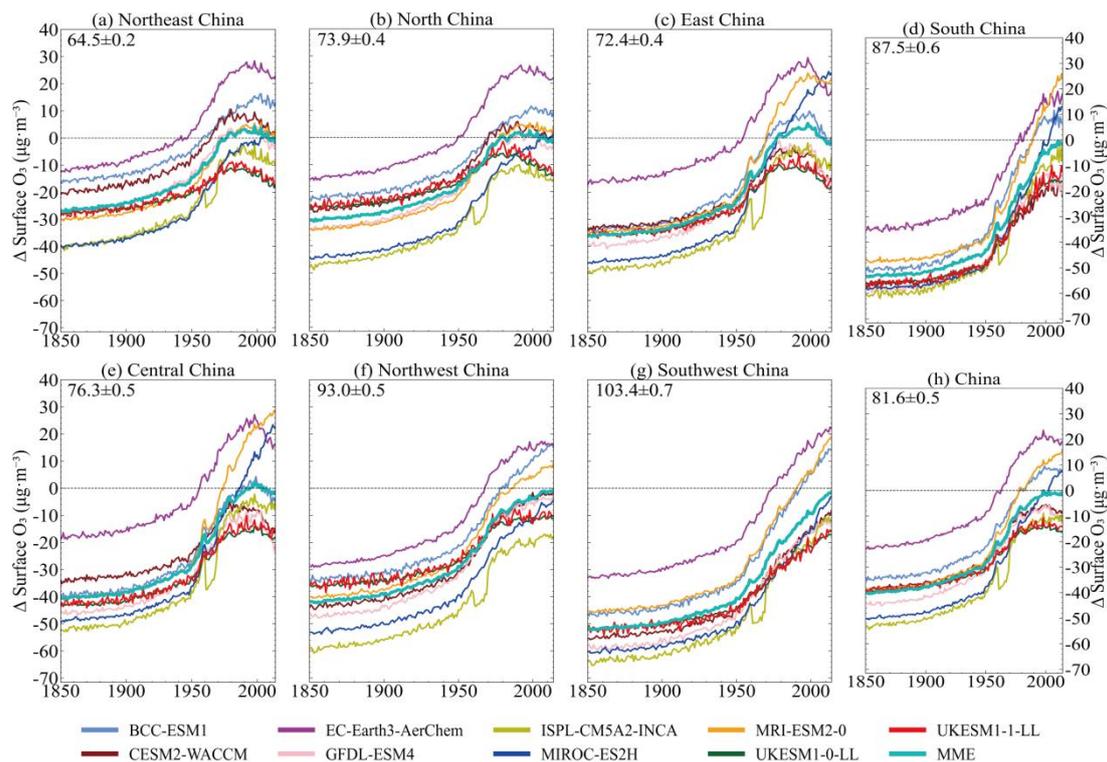


Figure 10. Changes in the China and sub-regions annual mean surface O₃ concentrations from the pre-industrial period to present day, relative to a 2015–2023 mean value, across nine CMIP6 models and MME. The multi-model annual mean 2015–2023 surface O₃ concentrations (± 1 SD) are shown in the top left of each panel.

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The simulated historical changes in surface O₃ concentrations across China show substantial regional and inter-model variations. For instance, UKESM1-0-LL simulates the smallest increase (most notably in Northeast China), whereas MIROC-ES2H simulates the largest (particularly in Central China), followed by MRI-ESM2-0 and BCC-ESM1. Although UKESM1-0-LL shows a minimal surface O₃ response, it projects a larger historical increase in tropospheric O₃ burden compared to other CMIP6 models (Griffiths et al., 2021). Moreover, the simulated historical changes in surface O₃ from UKESM1-0-LL are comparable to those from the HTAP_param (an emission-only-driven model), indicating that the magnitude of change simulated by UKESM1-0-LL is similar to that solely from changes in precursor emissions (Turnock et al., 2020). This implies that UKESM1-0-LL may exhibit relatively weak contributions from non-emission-driven processes (e.g., chemistry-climate interactions) compared with other CMIP6 models.

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Moreover, the simulated historical changes in surface O₃ from UKESM1-0-LL are comparable to those from the HTAP_param (an emission-only-driven model), indicating that the magnitude of change simulated by UKESM1-0-LL is similar to that solely from changes in precursor emissions (Turnock et

585

al., 2020). This implies that UKESM1-0-LL may exhibit relatively weak contributions from non-emission-driven processes (e.g., chemistry-climate interactions) compared with other CMIP6 models.

590 Central China exhibits the largest discrepancies in historical surface O₃ changes among the CMIP6 models, with the maximum inter-model difference reaching 29.6 μg·m⁻³. This pronounced inter-model spread can be understood in light of previous findings. As highlighted by Turnock et al. (2020), such variability in CMIP6 surface O₃ responses primarily arises from two factors: (i) differences in the simulated 1850 baseline O₃ concentrations, and (ii) differences in regional O₃ trend rates, reflecting
595 model-dependent chemical sensitivities to precursor emissions, particularly the nonlinear O₃-NO_x relationship. These mechanisms provide a consistent explanation for why models such as MIROC-ES2H simulate much stronger historical increases in surface O₃ than models like UKESM1-0-LL. Importantly, inter-model differences in chemical sensitivity extend beyond the response to NO_x alone, encompassing the treatment of other precursors and, critically,
600 aerosol-chemistry interactions. In Central China, precisely where the inter-model spread is greatest, large historical changes in PM_{2.5} concentrations (Su et al., 2022) may further modulate O₃ formation by influencing heterogeneous radical loss on aerosol surfaces. This aerosol modulation effect is likely most pronounced during seasons characterized by high PM_{2.5} loadings and weak photochemical activity (e.g., winter), when reduced solar radiation and enhanced atmospheric stability may amplify the impact
605 of aerosol-related processes on surface O₃.

In summary, the substantial inter-model spread in historical surface O₃ trends over China since 1850 reflects a combination of uncertainties associated with the pre-industrial baseline state and model-dependent chemical sensitivities. These sensitivities extend beyond precursor emissions alone, encompassing differences in chemical mechanisms and aerosol-chemistry interactions, which can
610 further modulate regional O₃ responses. This highlights the need for future multi-model attribution and constraint-based approaches to explicitly disentangle baseline-related uncertainty from process-level differences.

5 O₃ from present day to 2100

5.1 Projected trends under different SSP scenarios

615 Figure 11 shows the future changes in surface O₃ concentrations across China and its sub-regions under different CMIP6 scenarios (relative to the 2015–2023 mean). Overall, it is projected that by 2100, surface O₃ concentrations in China will decrease in most scenarios, with sub-regions responding similarly to the national trend, though with varying magnitudes (Figure 12). In the Tier 1 experiment, under the SSP1-2.6 scenario, which involves low radiative forcing, strong climate mitigation, and significant air pollution reduction, Surface O₃ concentrations in China are projected to decrease relative to the 2013–2024 annual mean, by $12.6 \pm 3.1 \mu\text{g}\cdot\text{m}^{-3}$ (± 1 SD of the MME), corresponding to a reduction of approximately 16% by 2050, and by $25.3 \pm 7.2 \mu\text{g}\cdot\text{m}^{-3}$, corresponding to a reduction of approximately 32% by 2100. Due to substantial reductions in precursor emissions, a decrease in CH₄ concentrations, and relatively small climate changes, under this scenario, surface O₃ concentrations in all sub-regions also show significant declines. Projections indicate that by 2100, surface O₃ concentrations in the Southwest and South China regions will decrease by more than $30 \mu\text{g}\cdot\text{m}^{-3}$, while in East China, which experiences the smallest reduction, O₃ concentrations will still decrease by nearly $20 \mu\text{g}\cdot\text{m}^{-3}$. For the medium forcing SSP2-4.5 scenario, it is projected that by 2100, the annual mean surface O₃ concentration in China will decrease by $13.6 \pm 7.2 \mu\text{g}\cdot\text{m}^{-3}$, corresponding to a reduction of 17%. Meanwhile, the projections indicate that the annual mean surface O₃ concentrations in all sub-regions of China will experience a slight increase around 2055 relative to the 2015–2023 mean, after which they begin to decrease. The most significant decrease occurs in South China, where it may reach $24.5 \mu\text{g}\cdot\text{m}^{-3}$ by 2100. This behavior is primarily driven by enhanced control of precursor emissions, relatively moderate climate changes, and variations in CH₄ concentrations. This mid-century rise is consistent with previous CMIP6 assessments, which show that under SSP2-4.5, precursor emissions decline only gradually while climate warming and photochemical activity increase rapidly before mid-century. As a result, climate-driven ozone enhancement can temporarily offset emission-driven declines, leading to a short-term increase around 2055 (Gidden et al., 2019; Turnock et al., 2020). In the SSP3-7.0 scenario, due to weak climate mitigation and weak air pollutant reduction, the annual mean surface O₃ concentration in China is projected to increase by $8.4 \pm 2.0 \mu\text{g}\cdot\text{m}^{-3}$ by 2050, and increase by $13.9 \pm 4.0 \mu\text{g}\cdot\text{m}^{-3}$ by 2100, with an increase of 17%. In this scenario, the annual mean

surface O₃ concentrations in all seven sub-regions show an upward trend, with the largest increase in East China, where the concentration is expected to rise by $19.3 \pm 6.9 \mu\text{g}\cdot\text{m}^{-3}$ by 2100, with an increase of 27%. Although emissions of O₃ precursors such as NO_x are projected to start decreasing around 2040 (Figure S3), the surface O₃ concentrations in all sub-regions continue to increase, indicating the importance of changes in chemical composition, increasing CH₄ concentrations, and climate change in the simulation of surface O₃ under the SSP3-7.0 scenario (Turnock et al., 2020; Young et al., 2013; Li et al., 2019). Additionally, the projected differences among CMIP6 models are most pronounced over Central and East China, with secondary hotspots of model spread also evident over Southwest and South China, indicating some divergence in the model simulations of O₃ across these regions.

In the SSP3-7.0-lowNTCF scenario, additional measures targeting non-methane short-lived climate forcers (SLCFs), including aerosols and ozone precursors, are implemented on top of the baseline SSP3-7.0 scenario. These measures are primarily aimed at improving air quality, without additional CO₂ or long-lived greenhouse gas mitigation (Allen et al., 2020). Reductions in SLCFs may slightly affect climate through changes in aerosol radiative effects. Consequently, the projections show that under the SSP3-7.0-lowNTCF scenario, the increase in surface O₃ concentrations in China is slower than in the SSP3-7.0 scenario, with relatively lower concentrations. By 2050, the annual mean surface O₃ concentration in China is projected to increase by only $5.8 \pm 1.5 \mu\text{g}\cdot\text{m}^{-3}$, and by 2100, it will increase by $4.9 \pm 2.0 \mu\text{g}\cdot\text{m}^{-3}$, representing a 6% increase. In this scenario, by 2100, surface O₃ concentrations in China and most of its sub-regions are expected to return to or be close to the 2015–2023 levels (especially in the Northwest China), showing a significant improvement in surface O₃ pollution compared to the SSP3-7.0 scenario. However, compared to other regions of the world, the additional reduction in precursor emissions under the SSP3-7.0-lowNTCF scenario has a relatively small impact on improving surface O₃ pollution in China. This is mainly due to the increase in surface O₃ concentrations in eastern China (especially in Central and Eastern China). This increase is caused by a slight rise in NMVOCs emissions and a reduction in O₃ titration due to a significant decrease in NO_x emissions (Turnock et al., 2020). Additionally, the decrease in PM_{2.5} concentrations under the SSP3-7.0-lowNTCF scenario leads to a reduction in the heterogeneous loss of free radicals, which may also contribute to the rise in surface O₃ concentrations (Li et al., 2019).

670 In the SSP5-8.5 scenario, characterized by high radiative forcing, weak climate mitigation, and weak air pollutant emission reductions, the annual mean surface O₃ concentration in China is projected to increase by 6.3±1.6 μg·m⁻³ by 2050.

In the SSP5-8.5 scenario, characterized by high radiative forcing, weak climate mitigation, and only limited air pollutant controls (Rao et al., 2017), the annual mean surface O₃ concentration in China is projected to increase by 6.3±1.6 μg·m⁻³ by 2050. However, by 2100, the surface O₃ concentration is expected to decrease by 3.4±2.9 μg·m⁻³ relative to the 2013–2024 mean, a reduction of approximately 4.2%. The projected changes in surface O₃ concentrations for the sub-regions in this scenario are similar to those in the SSP3-7.0-lowNTCF scenario (with a correlation of up to 0.7), despite differences in the underlying emissions and radiative forcing between the scenarios. By 2050, the surface O₃ concentration in most sub-regions will increase slightly faster in the SSP5-8.5 scenario than in the SSP3-7.0-lowNTCF scenario (except for Northeast and North China), but slower than in the SSP3-7.0 scenario (except for Central and East China). This may be attributed to the different changes in CH₄ emissions under different scenarios. Additionally, more CMIP6 model data are available for the SSP3-7.0 scenario (9 models) compared to the SSP5-8.5 scenario (4 models), which may also influence the MME response.

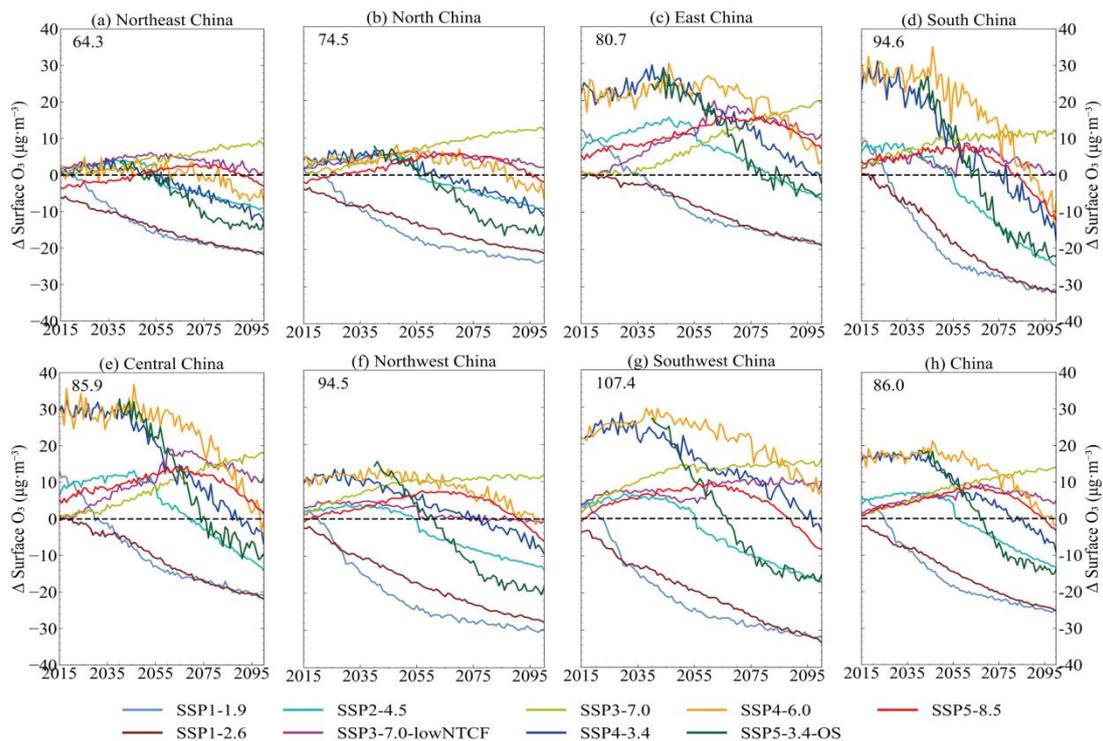
Under the Tier 2 experiment, the SSP1-1.9 scenario, characterized by the strongest climate mitigation and the most stringent air pollutant controls, leads to substantial reductions in surface O₃ levels over China. By 2050, the annual mean surface O₃ concentration is projected to decrease by 16.6±7.1 μg·m⁻³, and further decline to 25.3±9.5 μg·m⁻³ by 2100, corresponding to an overall reduction of approximately 32%. This pronounced decline is primarily driven by the large-scale and coordinated reductions in NO_x and VOCs under this scenario, which markedly suppress the photochemical production of O₃.

Meanwhile, although climate-induced changes and concurrent PM_{2.5} reductions can also influence O₃ through complex atmospheric chemical and physical processes, the dominant effect of precursor emission reductions ultimately leads to a substantial overall improvement in surface O₃ air quality under SSP1-1.9.

Under the SSP4-3.4 and SSP4-6.0 scenarios, which are characterized by regionally uneven climate mitigation and air pollutant reductions, the surface O₃ concentration in China is projected to increase by 13.5±1.3 μg·m⁻³ and 18.3±1.4 μg·m⁻³ by 2050, respectively. By 2100, the concentrations under these scenarios are expected to decrease by 9.2±8.3 μg·m⁻³ and 1.6±5.9 μg·m⁻³, respectively. In contrast,

700 SSP5-3.4-OS is an overshoot scenario, its radiative forcing pathway is designed to temporarily exceed the $3.4 \text{ W}\cdot\text{m}^{-2}$ level mid-century before declining back to that target by 2100. This pathway involves substantial late-century mitigation efforts, which also lead to reductions in air pollutant emissions as part of the broader deep-decarbonization strategy. Under this scenario, surface O_3 increases by $12.6 \pm 1.6 \mu\text{g}\cdot\text{m}^{-3}$ by 2050, comparable to the responses under the moderate-mitigation pathways. However, following the overshoot and the subsequent implementation of stringent mitigation, O_3 concentrations decline markedly by 2100, with a projected reduction of $13.9 \pm 11.7 \mu\text{g}\cdot\text{m}^{-3}$, the largest decrease among the three scenarios. This comparison highlights that although near-term O_3 increases across all three scenarios due to the combined influences of climate change and evolving precursor emissions, sustained and ambitious emission controls, particularly those implemented after an overshoot, as in

710 SSP5-3.4-OS, can ultimately produce substantial long-term reductions in surface O_3 , improving air quality and mitigating associated climate impacts.



715 **Figure 11. Future China and sub-regions changes in annual mean surface O_3 under different CMIP6 SSP scenarios. The dashed black line represents the curve of the difference at zero. For the Tier 2 scenarios (SSP4-3.4, SSP4-6.0, and SSP5-3.4-OS), O_3 data are available only from MRI-ESM2-0. As MRI-ESM2-0 simulates higher O_3 over most regions of China compared to the MME, these scenarios show larger initial deviations in the national panel and in some regions. The multi-model regional mean for 2015–2023 is shown in the top-left corner of each panel.**

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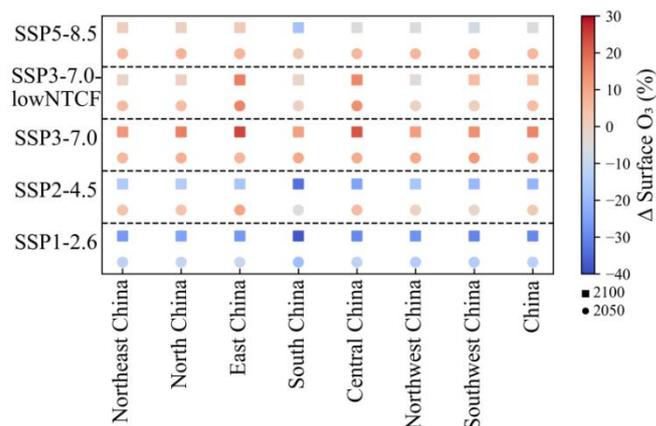


Figure 12. Percentage change in 2050 (circles) and 2100 (squares), relative to 2015, for annual mean of O₃ across China and sub-regions in the four Tier 1 future CMIP6 scenarios and the SSP3-7.0-lowNTCF scenario.

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In summary, projections from the CMIP6 models suggest that mitigating surface O₃ pollution across China will require not only reducing greenhouse gas emissions to moderate future climate change but also implementing enhanced controls on emissions of O₃ precursors (including CH₄). Under scenarios with pronounced climate change impacts, such as SSP3-7.0 and SSP5-8.5, the stringency of controls on key O₃ precursors, particularly NMVOCs and NO_x, beyond 2050 is expected to lead to divergent regional responses in long-term surface O₃ trends.

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5.2 Model spread and its drivers under the SSP3-7.0 scenario

Since the number of available CMIP6 models is the greatest under the SSP3-7.0 scenario (Table 1), this paper conducts a comparative analysis of the changes in surface O₃ concentrations across China and its sub-regions under the SSP3-7.0 scenario, aiming to identify the potential causes of model discrepancies. Figure 13 illustrates the changes in the annual mean and seasonal mean surface O₃ concentrations in China and its sub-regions for the years 2050 (2045–2055 mean) and 2095 (2090–2100 mean) relative to the 2015–2023 mean baseline, based on different CMIP6 models under the SSP3-7.0 scenario. O₃ is not directly emitted into the troposphere but is produced through photochemical oxidation of CO, CH₄, and NMVOCs in the presence of NO and NO₂. The abundance of tropospheric O₃ is determined by its budget, which includes chemical production, stratospheric transport, chemical loss, and deposition to the surface (Lelieveld and Dentener, 2000), and the intensity of these processes is highly sensitive to current climate conditions and the emissions and distribution of O₃ precursors (including NO_x,

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745 NMVOCs, CH₄, etc.). Therefore, this study further analyzes the correlation between future annual mean surface O₃ concentrations under the SSP3-7.0 scenario and other variables, including CH₄ concentrations, Near-Surface Air Temperature (TAS), NO_x concentrations, total emissions of NMVOCs and BVOCs (Figure 14).

It can be observed that surface O₃ concentrations predicted by different CMIP6 models under the
750 SSP3-7.0 scenario exhibit significant regional discrepancies. In particular, in Central China, the O₃ concentrations predicted by MRI-ESM2-0 and EC-Earth3-AerChem are nearly twice as high as those predicted by UKESM1-0-LL and GFDL-ESM4 (Figure S2). The lower annual mean O₃ concentrations in Central China for UKESM1-0-LL and GFDL-ESM4 are primarily attributed to higher NO_x emissions under the SSP3-7.0 scenario. In this region, NO_x emissions are approximately 2–3 times
755 higher than those in the other two models (Figure S2), which likely triggers NO_x titration and results in lower simulated surface O₃ concentrations. These inter-model differences in NO_x emissions arise because CMIP6 models use emission inputs derived from different source datasets and processing chains. Each modeling center applies its own downscaling, sectoral allocation, and harmonization procedures to the SSP scenarios, which can lead to notable regional discrepancies even when following
760 the same narrative. In contrast, in MRI-ESM2-0 and EC-Earth3-AerChem, NO_x titration is rare during DJF. In addition, EC-Earth3-AerChem exhibits higher simulated CH₄ concentrations (Fig. S4).

Although CH₄ forcing is prescribed and harmonized across CMIP6 models, inter-model differences in simulated CH₄ concentrations and their impacts on surface O₃ can still arise from differences in chemical mechanisms, particularly the representation of OH radical fields that control CH₄ lifetime.
765 Higher CH₄ levels in EC-Earth3-AerChem therefore enhance tropospheric background O₃ production, contributing to its relatively higher simulated O₃ concentrations in Central China. These discrepancies highlight that, although the driving factors related to O₃ changes (such as climate change and pollutant emissions) are crucial in all models (Figure 14), the differences in precursor emissions (NO_x and CH₄) and chemical process responses between models in future scenarios with significant climate change
770 have a substantial impact on regional O₃ concentration predictions.

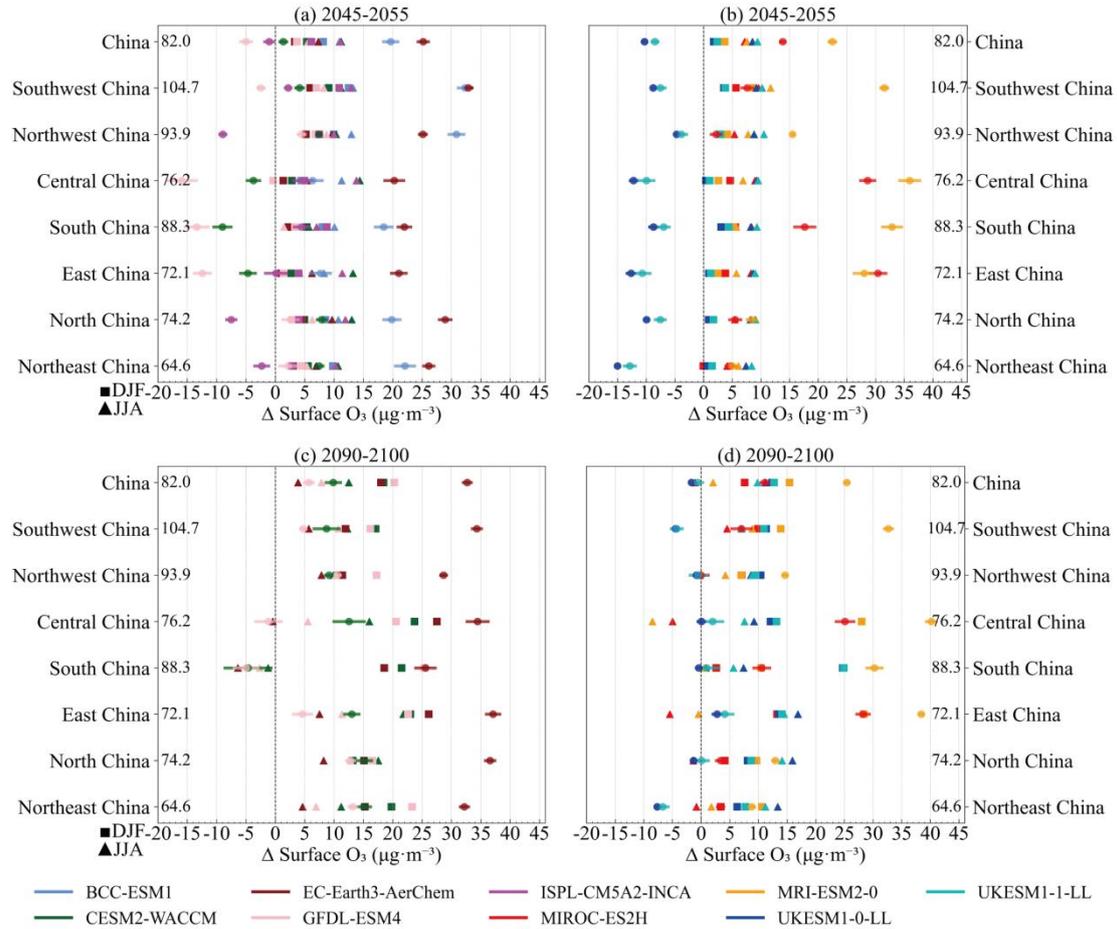


Figure 13. Changes in the annual and seasonal mean surface O₃ in China and its sub-regions, relative to the 2015–2023 mean, for the SSP3-7.0 scenario used in CMIP6. Each coloured circle represents the annual mean response for an individual model in (a) and (b) for 2045–2055, and in (c) and (d) for 2090–2100, with the coloured bars showing the SD across the annual mean. The seasonal mean responses for DJF and JJA, averaged over the relevant 10-year periods, are shown by squares and triangles, respectively. The multi-model regional mean for the 2015–2023 period is shown on the left of each panel.

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IPSL-CM5A2-INCA (predictions extending only to 2055) under the370 scenario, which projects that by 2050, surface O₃ concentrations in northern China (including Northeast, North, and Northwest China) will be lower than the 2015–2023 mean, with the most significant decrease expected in North China, where O₃ concentrations are projected to drop by approximately 10%. IPSL-CM5A2-INCA (ECS; 3.6 K) is a model with moderate equilibrium climate sensitivity, showing a moderate response to global temperature increases caused by greenhouse gases. The model simulates relatively high BVOCs emissions during the 2015–2023 period (covering a broader range of BVOCs types), with emissions approximately 4–8 times higher than those of other models (Figure S5), and these emissions have shown a consistent upward trend. However, despite the increase in BVOCs emissions, this model

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simulates relatively low NO_x concentrations and surface TAS, resulting in a smaller increase in O₃ concentrations, and even a decrease in some regions. This indicates that differences in the magnitude of climate change and O₃ precursor (NO_x) variations, as well as the different ways these factors are coupled in different CMIP6 models, lead to significant differences in the response of the models to BVOCs emissions. Such differences could directly influence future surface O₃ changes, particularly in localized regions.

795 CESM2-WACCM and GFDL-ESM4 under the SSP3-7.0 scenario, on the other hand, predict that by 2050, surface O₃ concentrations in southern China (including East, Central, and South China) will be lower than the 2015–2023 annual mean, with the most significant decrease observed in South China, where O₃ concentrations are projected to drop by 9% and 13% in the two models, respectively. Firstly, CESM2-WACCM (ECS; 4.7 K) and GFDL-ESM4 (ECS; 4.4 K) exhibit higher climate sensitivity, 800 meaning that their projected temperature increase and water vapor increase are more significant.

Higher temperatures and water vapor content facilitate the generation of OH radicals, which in turn accelerate O₃ destruction reactions (Wild et al., 2020). In tropical and subtropical regions, where both temperature and water vapor are already high, the presence of these factors may further enhance the consumption of O₃ by OH radicals, a phenomenon particularly evident in South China. Secondly, both 805 models tend to simulate lower BVOCs emissions (Figure S5), with GFDL-ESM4 showing the lowest and virtually unchanged BVOCs emissions under the SSP3-7.0 scenario, which may reduce the formation of O₃. Furthermore, the pollutant emissions and atmospheric chemical processes in southern China differ from those in the north. The southern regions are likely more dependent on photochemical reactions, which are more active under higher temperatures and stronger solar radiation conditions.

810 Therefore, CESM2-WACCM and GFDL-ESM4 may simulate a greater number of photochemical reactions, further accelerating O₃ decomposition.

Under the SSP3-7.0 scenario, both UKESM1-0-LL and UKESM1-1-LL project decreases in surface O₃ concentrations across China and its sub-regions by 2050 relative to the 2015–2023 annual mean, with the largest reductions of 23% and 20% occurring in Northeast China. Although a moderate increase is 815 projected by 2095, O₃ concentrations remain below the 2015–2023 baseline in most regions, except East China. Compared to other CMIP6 models, UKESM1-0-LL (ECS; 5.4 K) and UKESM1-1-LL (ECS; 4.2 K) exhibit higher climate sensitivities. The elevated temperatures (Figure S7) and altered climatic conditions in these models likely enhance O₃ degradation, contributing to the generally lower

O₃ concentrations in their simulations. Moreover, UKESM1-0-LL simulates higher atmospheric NO_x levels, promoting NO_x titration that suppresses O₃ formation. This model also projects higher NMVOCs emissions (Figure S6). Under high-NO_x conditions, the interplay between NO_x and NMVOCs can modify photochemical O₃ production pathways, further inhibiting net O₃ formation (Jiménez and Baldasano; Xing et al., 2011). To further investigate this mechanism, we constructed a two-dimensional framework based on ground observations, analyzing O₃ distribution across China within the NO₂-NMVOCs space (Figure 15). Results indicate that the highest O₃ levels occur under moderate NO₂ (20–40 μg·m⁻³) and elevated NMVOCs (>500 t·month⁻¹) conditions, reflecting a typical VOC-limited O₃ formation regime. In such environments, abundant NMVOCs coupled with relatively low NO_x levels promote efficient photochemical O₃ production. However, when NO₂ concentrations reach 60–120 μg·m⁻³, O₃ decreases significantly even at intermediate-to-high NMVOCs, indicating strong inhibition of O₃ formation by excess NO_x. It is important to note that the O₃ formation regime depicted in Figure 15 is derived from present-day observations and should be regarded as a diagnostic reference rather than a fixed projection of future atmospheric chemistry. CMIP6 multi-model simulations under SSP3-7.0 project that NO_x emissions increase from 2015 and peak around 2035 before gradually declining to near 2015 levels by 2065 (Figure S3), while NMVOCs emissions continue a modest upward trend throughout the century (Figure S6). Based on these projections, our analysis suggests that many regions in China could remain in or near the high-NO_x suppression regime during the first half of the 21st century (around the NO_x emission peak), where O₃ formation could be inhibited by excessive NO_x. As NO_x emissions decline thereafter, the chemical environment is projected to gradually shift towards a regime more sensitive to NMVOCs, potentially approaching VOC-limited conditions, where O₃ production becomes increasingly dependent on NMVOCs levels. Regional variations in the relative reduction rates of NO_x and NMVOCs are expected to further influence these transitions. These potential shifts highlight the need for adaptive, region-specific emission-control strategies that account for evolving chemical environments.

The projections of annual mean surface O₃ concentrations for China and its sub-regions under the SSP3-7.0 scenario by BCC-ESM1 (ECS; 4.0K, predictions extending only to 2055), EC-Earth3-AerChem (ECS; 3.0K), MIROC-ES2H (ECS; 3.6K), and MRI-ESM2-0 (ECS; 5.4K) show significant consistency, with O₃ concentrations in 2050 and 2095 both being higher than the 2015–2023 mean. Among these models, MIROC-ES2H and MRI-ESM2-0 exhibit higher climate sensitivity,

850 although their projected NO_x and CH_4 concentrations are relatively low (Figure S3–S4), they still tend to predict larger increases in O_3 compared to the other models, with the most significant O_3 increase observed in southern China (including East China, Central China, and South China). In contrast, BCC-ESM1 and EC-Earth3-AerChem predict more noticeable increases in O_3 concentrations in northern China (including Northeast, North, Northwest, and Southwest China). EC-Earth3-AerChem, with a relatively low ECS, simulates lower NO_x concentrations in China and its sub-regions, while its CH_4 concentrations are the highest among the models (Figure S4). Under low NO_x conditions and a weak NO_x titration effect, an increase in CH_4 promotes O_3 formation, leading to O_3 accumulation and a more pronounced increase in simulated O_3 concentrations in the model. BCC-ESM1, on the other hand, tends to simulate higher NO_x concentrations and lower TAS (Figure S3 and S7), which contributes to the more noticeable O_3 increase predicted by this model.

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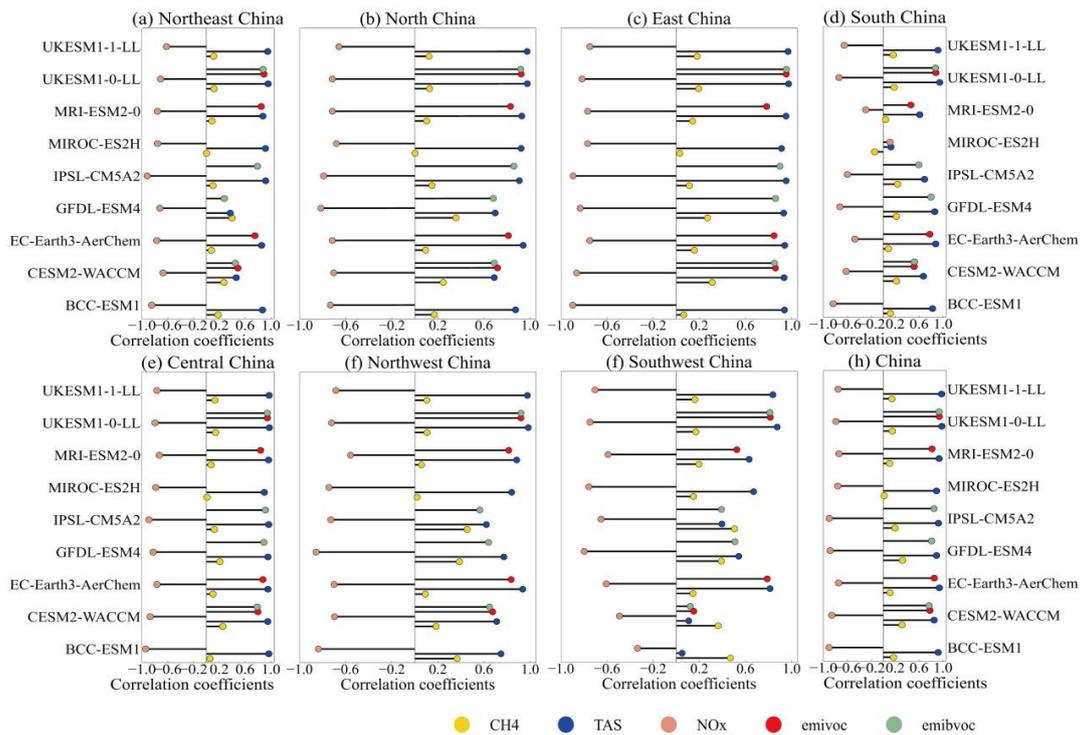


Figure 14. Correlation coefficients calculated when comparing future annual mean surface O_3 concentrations against individual variables of surface CH_4 concentrations, temperature at Surface (TAS), emissions of NMVOCs and BVOCs, NO_x ($\text{NO} + \text{NO}_2$) concentrations and from individual CMIP6 models over the period 2015 to 2100 in the SSP3-7.0 scenario.

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The seasonal responses of different models under the SSP3-7.0 scenario also show variations across sub-regions. Most models predict that surface O_3 concentrations increase more in JJA than in DJF.

However, some regions exhibit a decreasing trend in O₃ concentrations during JJA, which aligns with
870 the findings of Turnock et al. (2020).

Figure 14 reveals that under the SSP3-7.0 scenario, the correlations between surface O₃ and its driving
variables show both consistent patterns across CMIP6 models and region-dependent variability. The
robust negative O₃-NO_x correlation across all models quantitatively confirms the dominance of NO_x
titration in China over the study period (2015–2100). However, the magnitude of this correlation varies
875 substantially ($r = -0.92$ in BCC-ESM1 over Central China and -0.11 in MIROC-ES2H over South
China). This spectrum of correlation strengths is a key diagnostic of inter-model differences in
representing O₃ photochemistry. Specifically, the stronger negative correlations indicate that these
models simulate a more extensive or persistent VOC-limited chemical regime under the SSP3-7.0
pathway. Consequently, the projected NO_x declines within these models trigger a more complete
880 relaxation of titration, resulting in a larger O₃ increase.

The consistently strong positive O₃-TAS correlation, with all models exceeding 0.85 over China as a
whole and in most sub-regions, underscores climate warming as a major and predominant driver of
future O₃ trends. A clear exception is the Southwest China, where most models simulate lower
correlations, reinforcing the interpretation that non-thermal controls (e.g., the complex terrain of the
885 Tibetan Plateau and international transport) play a unique role there. Against this backdrop of high
consensus, models like UKESM1-0-LL, which sustain correlations >0.9 across nearly all regions,
likely simulate particularly efficient temperature-dependent processes (e.g., in BVOCs emissions or
peroxy radical chemistry), thereby projecting an amplified warming effect on O₃.

Although a positive O₃-CH₄ correlation is a common feature across models, its highly variable strength
890 highlights key uncertainties in how they represent methane's role. The MIROC-ES2H is a prominent
outlier, simulating consistently weak and, in some regions even negative correlations ($r = -0.14$ in
South China). This, coupled with its atypical weak (or even positive) O₃-NO_x correlation in the same
region, strongly suggests that O₃ production in MIROC-ES2H is predominantly governed by the local
interplay of NO_x and VOCs, whose strong control effectively masks the influence of the global CH₄
895 background. In contrast, models like IPSL-CM5A2 ($r = 0.51$ in China) simulate a clear positive
correlation, indicating that their frameworks allow methane oxidation chemistry to play a substantive
and detectable role in future O₃ production.

The strong but model-dependent O₃-VOCs positive correlations provide further diagnosis of chemical regime representations. Models such as EC-Earth3-AerChem and UKESM1-0-LL show high sensitivity to both NMVOCs and BVOCs emissions (with correlations often >0.8), which is characteristic of a VOC-sensitive chemical environment. A particularly telling diagnostic is that in these models, O₃ correlates more strongly with VOC emissions than with near-surface NO_x concentrations. This pattern potentially reflects differences in the represented chemical nonlinearity of the VOCs-NO_x-O₃ system or the relative atmospheric lifetimes and transport of the precursor species.

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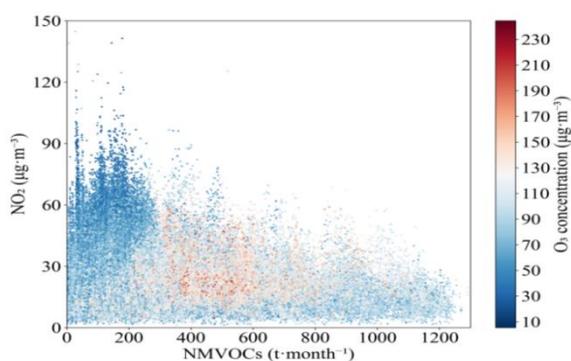


Fig. 15 Distribution of O₃ concentration in the NO₂-NMVOCs coordinate over China, based on 2015–2023 surface observations of NO₂ and O₃ and MME NMVOCs under the SSP3-7.0 scenario.

910 The differences in simulations between different CMIP6 models highlight the importance of further understanding how future O₃ concentrations will be influenced by the combined effects of pollutant emissions (especially with regard to the differences in how O₃ precursors, PM_{2.5}, and other factors are coupled and how chemical processes respond across models) and climate change. For example, in the Central China region, the prediction differences between models are highly significant, with some models predicting O₃ concentrations that could be twice as high as those of others. This discrepancy reflects the need for future research to focus more on model uncertainty in order to improve the accuracy of future air quality predictions.

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6 Summary and discussion

This study provides a comprehensive evaluation of surface O₃ over China using the CMIP6 multi-model ensemble, spanning present-day performance, historical evolution (1850–2014), and future projections under diverse scenarios. By synthesizing findings across these temporal scales, we

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offer an integrated perspective on model capabilities, key sources of uncertainty, and the implications for understanding past changes and future air quality. The principal insights are discussed below.

925 (1) The CMIP6 MME reproduces the observed spatial and seasonal pattern of surface O₃, with maxima in summer and over central China. However, a systematic underestimation prevails across most models and regions, particularly in eastern China. This bias is not random; rather, it tends to be modulated by environmental conditions: model error generally increases over urban surfaces and under high PM_{2.5} concentrations, revealing a complex, non-linear relationship especially in winter. Critically, these identified present-day biases are instructive for interpreting simulations of other periods. They point to 930 persistent structural model limitations in key processes, particularly photochemical mechanisms and aerosol-chemistry interactions. Consequently, historical or future periods characterized by environmental conditions analogous to those linked with high bias today are likely subject to similar directional biases in simulated O₃. Acknowledging and accounting for this transferable understanding of model bias can therefore help advance a more robust interpretation of both past changes and future 935 projections.

(2) The MME indicates a substantial increase of $39.3 \pm 14.4 \mu\text{g}\cdot\text{m}^{-3}$ in China's annual mean surface O₃ since 1850, with a marked acceleration post 1950. The large inter-model spread in this trend originates primarily from two factors highlighted in the literature: (i) differences in the simulated pre-industrial (1850) baseline concentrations, and (ii) divergent chemical sensitivities of O₃ production to emission 940 changes. This spectrum of sensitivity is clearly evident in our results: MIROC-ES2H exhibits high sensitivity and simulates the largest historical trend, whereas UKESM1-0-LL shows lower sensitivity, producing a trend most consistent with a direct, emission-driven response. The fact that the greatest model disparity occurs in Central China, a region also undergoing significant aerosol changes, further highlights the contribution of aerosol-chemistry interactions to the overall projection uncertainty.

945 (3) Future surface O₃ levels over China are governed by the complex interplay between air pollution control and climate mitigation. A pronounced decrease occurs under the strong co-mitigation scenario SSP1-2.6. In stark contrast, the high-forcing SSP3-7.0 pathway projects continuous O₃ increases in many regions. Our analysis reveals that the common projection of a mid-century O₃ peak followed by a decline can arise from two fundamentally different policy dominances: pathways where climate 950 mitigation objectives are central (SSP2-4.5), and pathways where improvements are primarily driven by stringent air quality controls even under high climate forcing (SSP5-8.5). This underscores a critical

insight: substantial O₃ reductions are achievable under divergent socioeconomic futures, but they demand either deep coupled climate-air quality governance or, alternatively, exceptionally strong dedicated air pollution policies in the absence of climate action.

955 (4) This uncertainty arises from fundamental differences in how models represent key chemical and climatic processes. Under SSP3-7.0, while models generally agree on the positive roles of climate warming (TAS), global background chemistry (CH₄), and precursors (NMVOCs), they diverge sharply in simulating the local efficacy of NO_x control, as indicated by inconsistent O₃-NO_x correlations. Such divergence underscores that the simulated O₃ response is not merely a function of emission inputs but
960 is critically dependent on each model's representation of complex, non-linear transition between chemical regimes (e.g., VOC-limited vs. NO_x-saturated), which can be further modulated by model-specific climate sensitivities and background emission levels. Consequently, the optimal strategy for future O₃ abatement, specifically, the relative priority and expected benefit of controlling NO_x versus managing other drivers like CH₄ or NMVOCs under a warming climate, remains highly
965 model-dependent. This poses a fundamental challenge for deriving robust, model-consistent policy insights, highlighting the need to interpret projections within the context of these underlying chemical mechanisms.

Overall, the results of this study are broadly consistent with conclusions from CMIP6-based global-scale analyses (Turnock et al., 2020; Griffiths et al., 2021; Zeng et al., 2022), while further
970 revealing more pronounced regional heterogeneity. Consistent with global studies, surface ozone concentrations over China decrease markedly under the strong mitigation scenario (SSP1-2.6), but exhibit overall or episodic increases under high-emission scenarios (SSP3-7.0 and SSP5-8.5). However, under the SSP3-7.0-lowNTCF scenario, which emphasizes reductions in short-lived climate forcers, surface O₃ over China still shows an increasing tendency, in clear contrast to the global mean response.
975 This discrepancy highlights the important role of regional chemical processes and emission structures in modulating ozone changes. Furthermore, this study finds that the magnitude of ozone changes over China is generally larger than the global average, accompanied by more pronounced spatial heterogeneity (Table S5). In terms of chemical mechanisms, the significant negative correlation between O₃ and NO_x over China—indicating the dominance of NO_x titration—is consistent with
980 established understanding for East Asia, whereas global-scale analyses suggest substantial regional heterogeneity in this relationship (Turnock et al., 2020). The positive correlations between surface O₃, CH₄, and TAS are also consistent with CMIP6-based global studies. In contrast, the relationships between O₃ and NMVOCs and BVOCs exhibit pronounced complexity and substantial inter-model

variability, which is consistent with findings from global multi-model analyses identifying O₃-VOC
985 sensitivity as one of the key sources of uncertainty in future ozone projections. Consequently, this
study not only confirms the applicability of the global scenario framework to the China region, but also
quantitatively characterizes the magnitude and uncertainty of ozone changes within key high-emission
regions, underscoring the importance of regionally refined process-based analyses in future air quality
assessments.

990 This study is based on the CMIP6 multi-model ensemble mean. The MME approach integrates
information from different models and mitigates the influence of individual model biases, yet it carries
certain limitations. First, the CMIP6 global models operate at coarse spatial resolutions (>100 km),
which limits their ability to resolve fine-scale urban features, emission heterogeneity, and local
pollution hotspots. Consequently, peak pollutant concentrations in highly urbanized regions may be
995 underestimated, and the results are therefore more suitable for analyzing regional-scale trends rather
than city-scale exposure assessments.

Second, the MME was calculated using an equal-weighted scheme, following the common
“one-model-one-vote” approach in model intercomparison studies. This method inherently assumes
model independence. It should be noted that some models (e.g., UKESM1-0-LL and UKESM1-1-LL)
1000 share similar atmospheric chemistry schemes, which may bias the ensemble toward specific model
families. Third, due to the varying data availability for different CMIP6 scenarios, the number of
contributing models differs across scenarios. Consequently, the uncertainty ranges (e.g., SD) of the
MME are not strictly comparable between scenarios. Despite these limitations, the main qualitative
conclusions of this study are robust, and the primary trends are not dominated by any single model.

1005 Future work could consider employing performance-based or independence-weighted ensemble
methods, or utilizing more comprehensive and balanced model ensembles, to better quantify
uncertainties arising from inter-model structural differences.

Data availability. The CMIP6 data can be accessed and downloaded at
1010 <https://aims2.llnl.gov/search/cmip6/> (last access: 8 September 2024). TAP data can be and downloaded
at <http://tapdata.org.cn/> (last access: 22 July 2024).

Author contributions. SL, HZ, QC, YC, ZW, QA and XW designed the study. XW and QA carried out

the data collection. SL, QC and YC carried out the data processing and analysis. SL, HZ, and ZW
1015 assisted with the interpretation of results. All co-authors contributed to writing and reviewing the
paper.

Competing interests. The authors declare that they have no conflict of interest.

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