



# 1 The 21st-century wetting inhibits growing surface ozone in Northwestern

# 2 China

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### ABSTRACT:

- 16 Previous studies have shown that surface air temperature (SAT) facilitates the formation of surface
- 17 ozone (O<sub>3</sub>), while relative humidity (RH) often inhibits ozone generation. However, the degree to
- 18 which O<sub>3</sub> may respond simultaneously to rising SATs and RH due to climate change remains less
- 19 understood. We conducted extensive atmospheric chemistry model scenario simulations to
- 20 investigate the impacts of long-term trends of humidification and warming on summer O<sub>3</sub>
- 21 concentrations in Northwestern China (NW) from 1998 to 2017, a period during which this region
- 22 experienced both warming and the most significant wetting trend in China. We found that the
- 23 summer mean O<sub>3</sub> level in NW increased by 19.9% during this time. The changes in meteorology
- 24 led to a reduction in O<sub>3</sub> levels, primarily due to humidification in NW, which counteracts the
- 25 warming-induced O<sub>3</sub> increase. We demonstrate that the wetting trend in NW will continue from
- 26 2019 to 2030 under a shared socioeconomic pathway (SSP) in conjunction with a representative
- 27 concentration pathway SSP5-RCP8.5, but will either cease or shift to drier conditions from 2019
- 28 to 2060 under the SSP2-RCP4.5. No significant responses of O<sub>3</sub> fluctuations to RH variations were
- 29 observed, partly due to manual intervention under the SSP2-RCP4.5.
- 30 **Keywords:** Ozone, Wetting, Warming, Northwestern China, Future projection





#### 1. Introduction

Atmospheric humidity is recognized as a crucial factor that directly and indirectly influences the formation and evolution of surface ozone (Ghazali et al., 2022; Han et al., 2011; Kerr et al., 2020). The relationship between atmospheric humidity and surface O<sub>3</sub> is complex. From a chemical standpoint, humidity can modify the rates of photochemical reactions, thereby affecting O<sub>3</sub> formation. Additionally, humidity plays a role in generating hydroxyl radicals (OH) in the atmosphere, which are vital for breaking down volatile organic compounds (VOCs) and other pollutants (Ding et al., 2023; Ma et al., 2021). These radicals can interact with O<sub>3</sub> precursors, impacting the rate of O<sub>3</sub> production. High humidity can also enhance aerosol formation, facilitating heterogeneous reactions. Depending on the specific chemical pathways involved, some of these reactions may either generate or deplete O<sub>3</sub> (Eck et al., 2020). From a physical perspective, humidity is critical for cloud and fog formation as well as precipitation. The latter promotes O<sub>3</sub> dissolution in water droplets and its removal from the atmosphere through wet deposition, particularly during precipitation events, which weakens solar radiation and disrupts the energy balance near the surface (Ding et al., 2021; Eck et al., 2020; Kleeman, 2008; Madden and Williams, 1978; Millstein and Harley, 2009).

It has been reported that O<sub>3</sub> levels increased by 30% over Northern China in summer and 50% over Eastern China in autumn during a dry year compared to a wet year, indicating that wet climate conditions do not support rising O<sub>3</sub> levels (Ding et al., 2023). Long-term ambient O<sub>3</sub> measurement data indicate that rising absolute air humidity resulted in lower near-surface O<sub>3</sub> concentrations at temperatures of 0–30°C (Belan and Savkin, 2019). Other studies examining the relationship between O<sub>3</sub> concentrations and atmospheric humidity have also identified negative correlations between O<sub>3</sub> levels and humidity (Li et al., 2022). Nevertheless, significant knowledge gaps regarding the relationship between humidity and O<sub>3</sub> persist (Li et al., 2021). Most previous studies utilized short-term or instantaneous O<sub>3</sub> concentrations to evaluate the associations between O<sub>3</sub> and atmospheric humidity (Gong et al., 2021; Li et al., 2021). Their long-term relationships still require clarification. Previous investigations of the connections between climate and O<sub>3</sub> primarily focused on the impact of air temperature, which often overshadows the effects of other meteorological variables (Wang et al., 2024). The extent to which atmospheric humidity could significantly influence the long-term O<sub>3</sub> trend remains uncertain, given that the synergistic effects of warming and wetting frequently occur in a warming climate. In this context, O<sub>3</sub> and RH trends in NW over





62 the past decades likely present a unique scenario to help bridge this knowledge gap. NW (Fig. S1), 63 located in the hinterland of the Asian-European continent, is Asia's largest arid and semi-arid 64 region, characterized by limited rainfall and vegetation (Koster et al., 2004; Yang et al., 2023). 65 Meteorological records indicate that from 1960 to 2013, precipitation in NW increased at a rate of 0.55 mm per year (Li et al., 2012; Liu et al., 2013; Yang et al., 2017). During this period, the air 66 temperature rose at a rate of 0.034 °C per year, exceeding the warming trends observed across all 67 68 of China (0.025 °C per year) and globally (0.013 °C per year). Although RH does not exhibit a 69 significant growth trend during this period, in the recent decade (2008-2017), it increased markedly 70 at a slope of 0.313 in NW and 0.381 in the three westernmost provinces of Xinjiang, Gansu, and 71 Qinghai within NW. Based on these records, NW appears to be entering a "warming and wetting" 72 phase, accompanied by a rise in precipitation, runoff, and lake levels (Nie et al., 2022; Peng and 73 Zhou, 2017; Shi et al., 2002; Yao et al., 2021). 74 The present study aims to address the knowledge gap in the complex and long-term 75 relationships between ozone pollution and atmospheric humidity, focusing on NW, where 76 increasing O<sub>3</sub> pollution, air temperature, and humidity have co-occurred (Peng and Zhou, 2017; 77 Yang et al., 2017), mainly since the early 2000s. This research provides valuable insights into the 78 broader context of air quality management and climate variability by enhancing our understanding

of the long-term relationship between ozone pollution and atmospheric humidity.

2. Method and data

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### 2.1 WRF-Chem Model Configuration

Since measured  $O_3$  concentration data in China is only available after 2013, we applied the three-dimensional Weather Research and Forecasting with Chemistry (WRF-Chem) version 3.7 model (http://www2.mmm.ucar.edu/wrf/users/wrf\_files/wrfv3.7/updates-3.7.html) to predict meteorology,  $O_3$  concentrations, and other relevant data in Northwestern and mainland China. The model domain spans the entire mainland China at a  $20 \text{ km} \times 20 \text{ km}$  resolution, extending vertically from the surface to the lower stratospheric ozone layer at an altitude of 50 hPa with 30 staggered vertical layers. This vertical segmentation facilitates a deepened comprehension of the  $O_3$  dynamics within the region, enabling the capture of its detailed behavior throughout the





91 atmospheric stratifications (Li et al., 2020; Zhang et al., 2020, 2022). Detailed physical and 92 chemical schemes contributing to O<sub>3</sub> formation in WRF-Chem and emission input followed 93 previous studies (Bei et al., 2018; Bossioli et al., 2016; Liao et al., 2015; Tie et al., 2009; Wild et 94 al., 2000) and are presented in Section S1 in the Supplement and Tables S1-S2. The soil NO, NO<sub>x</sub>, and HONO emissions have been considered a necessary process contributing to surface ozone 95 formation in recent years (Lu et al., 2021). However, given the lack of soil emission inventories 96 97 of these reactive nitrogen species in China and the world, the present modeling study did not take 98 these processes into consideration. The aerosol feedback mechanisms include the direct and 99 indirect effects, the both are considered in WRF-Chem. The model simulations with a nudging 100 option in WRF were conducted from June 1st to August 31st in each summer from 1998 to 2017, with a time-step of 30s. A spin-up time of three days was adopted before June 1<sup>st</sup> in each summer. 101 102 Further details are referred to Section S1 in the Supplement.

### 103 **2.2 Trend**

Considering relatively short data time series (1998-2017), the Mann-Kendall (MK) Test and Sen's slope (El-Shaarawi and Niculescu, 1992; Kendall, 1949; Mann, 1945) were used to examine the trend of O<sub>3</sub>, near-surface air temperature (SAT) at the 2m height, and relative humidity (RH, %) at the 2m height. The Mann-Kendall (MK) Test and Sen's Slope are both non-parametric statistical methods commonly used for analyzing trends in time series data. The MK Test helps determine if there is a significant trend, and Sen's slope quantifies the strength and direction of the trend (slope value).

# 2.3 O<sub>3</sub> Attribution to Meteorology and Emission

We calculated the fraction (percentage change, %) to account for  $O_3$  attribution to meteorology

and emission subject to different model scenarios. For example, we define the fraction

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$$F_{O3} = (C_{S3} - C_{S1}) / C_{S1} \times 100\%,$$
 (1)

as the attribution of  $O_3$  concentration to emissions, where  $C_{S1}$  and  $C_{S3}$  are the summer surface  $O_3$ 

air concentrations from the base (S1) and fixed meteorology (S3) scenarios.

#### 2.4 Data

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118 To initialize and prescribe boundary conditions in the WRF model from 1998 to 2017, 119 meteorological data were sourced from the ERA-Interim reanalysis from the European Centre for 120 Weather Medium-Range Forecast (ECMWF, 121 https://www.ecmwf.int/en/forecasts/datasets/reanalysisdatasets/era-interim). 122 Anthropogenic emissions were collected from the Emissions Database for Global 123 Atmospheric Research (EDGAR) version 4.3 on a spatial resolution of  $0.1^{\circ} \times 0.1^{\circ}$ , which was 124 available when we started this modeling investigation. A grid-mapping program implemented the 125 emissions into the WRF-Chem (Crippa et al., 2018; Mo et al., 2017). Biogenic emissions were 126 simulated using MEGAN version 2.1 (Guenther et al., 2006, 2012). A recent study suggests that 127 temperature can alter precursor emissions (Pfannerstill et al., 2024). However, since available 128 emission inventories worldwide, including the widely-used EDGAR inventory, did not account 129 for the influence of meteorology, typically the air temperature, on emissions but only considers 130 emissions from anthropogenic and natural sources, the emissions input in the present modeling 131 might be subject to additional uncertainties. The reduction of these uncertainties will depend on 132 improved inventories that take the meteorological effect into account. 133 We have replaced WRF land-use change (LUC) data with Chinese land use/land cover remote 134 sensing monitoring database provided by the Data Center for Resources and Environmental 135 Sciences, Chinese Academy of Sciences (RESDC) (http://www.resdc.cn), which significantly 136 improved China's LUC in our investigation of the effect of urbanization on O<sub>3</sub> pollution in China 137 (Zhang et al., 2022). The new LUC data has a spatial resolution of 1 km × 1 km, which is 138 extrapolated to the WRF-Chem grid cells ( $20 \text{ km} \times 20 \text{ km}$ ). 139 Considering that the warming and wetting trends in NW might persist in the coming decades, 140 we also projected future O<sub>3</sub> and meteorology in 2019, 2030, and 2060 in the WRF-Chem model 141 framework and simulations, respectively. We collected future anthropogenic emission data from 142 the Dynamic Projection Model for Emissions in China (DPEC Inventory - MEICModel, 143 http://meicmodel.org.cn), with a spatial resolution of  $0.25^{\circ} \times 0.25^{\circ}$  longitude and latitude (Cheng 144 et al., 2021, 2023; Tong et al., 2020). The meteorological data was collected from Shared 145 Socioeconomic Pathways (SSP) and Representative Concentration Pathways (RCP) climate 146 scenarios under the CMIP6 framework (https://aims2.llnl.gov/search/cmip6/), with the MPI-147 ESM1-2-HR model (Eyring et al., 2016). Compared with other models, MPI-ESM1-2-HR has 148 higher resolution and can simulate the complex interactions within the climate system, including



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all climate scenarios (SSP2-RCP4.5 and SSP5-RCP8.5) targeted by this study, and has been widely used in WRF simulations.

#### 2.5 Model Evaluations

Rigorous comparisons between simulated  $O_3$  concentrations and measured data were conducted and presented in Figs. S2-S7 and Section S2 in the Supplement. Overall, the simulated  $O_3$  concentrations agree reasonably well with measured data, demonstrating a high level of reliability of modeling results.

# 2.6 Model Scenario Setup

We established several model scenarios to distill signals of meteorological conditions, primarily SAT and RH, in modeled O<sub>3</sub> concentrations. Detailed configurations are provided in Table 1. The first scenario (S1) serves as the baseline, incorporating summer-specific meteorological variables and emissions from 1998 to 2017 into WRF-Chem simulations, and is thus considered a realistic scenario. In the second scenario (S2), we fixed emissions from 1998 throughout the period from 1998 to 2017, aiming to assess the significance of the meteorological effect on O<sub>3</sub> variation. In the third scenario (S3), we fixed meteorology from 1998 during the same period, thereby emphasizing the importance of emissions in  $O_3$  variation. To evaluate the sensitivity of scenario S3, we also conducted an additional WRF-Chem run by fixing SAT and RH only in 1998 (scenario S4) and compared the modeled O<sub>3</sub> concentrations with those simulated in scenario S3 for 2004. The results indicate minor differences between model scenario S3 and the results from the fixed SAT and RH model run (Fig. S8). By calculating the differences and fractions, we can discern the contributions of meteorology and emissions to O<sub>3</sub> formation and destruction. We also established two additional model scenarios to project the responses of O<sub>3</sub> concentrations to future meteorology and emissions in NW based on CMIP6 pathways SSP2-RCP4.5 and SSP5-RCP8.5, specifically for 2019, 2030, and 2060, respectively. Detailed descriptions of these two pathways and the corresponding data are presented in Section S3 in the Supplement.

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#### **Table 1.** Model scenarios.

Scenario	Simulation period	Meteorology	Emission	LUC
S1	1998-2017	Varied	Varied	Varied
S2	1998-2017	Varied	Fixed (1998)	Varied
S3	1998-2017	Fixed (1998)	Varied	Varied
S4	2004	Only fixed SAT and RH (1998)	Varied	Varied
S5	1998-2017	Varied	Varied	Fixed (1998)
SSP2-4.5	2019, 2030, 2060	Varied	Varied	Varied
SSP5-8.5	2019, 2030, 2060	Varied	Varied	Varied

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### 3. Results and Discussion

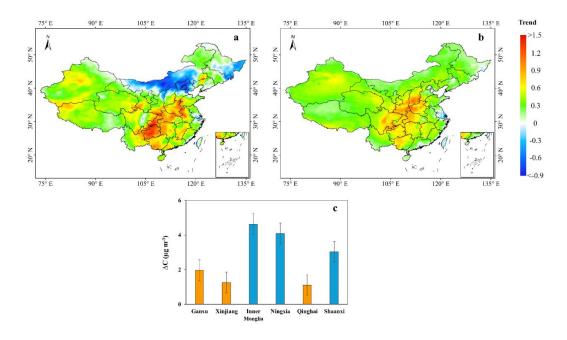
## 3.1 Summer Ozone Trends

Figures 1a and 1b present MK-Test statistics and Sen's slope estimated temporal trends of summer O<sub>3</sub> concentrations from 1998 to 2017 under scenario S1 (base scenario) and S3 with fixed meteorology in China. In the S1 scenario (Fig. 1a), O<sub>3</sub> concentrations exhibit statistically significant declining trends in the northernmost regions, spanning from Northeastern Xinjiang to Northeastern China, with the most significant decline occurring in Inner Mongolia. Increasing O<sub>3</sub> concentrations are observed along Tianshan Mountain in Xinjiang, western Tibet, and central China. The fixed meteorology scenario (S3, Fig. 1b) shows a monotonically increasing trend across China, indicating continuously rising O<sub>3</sub> levels for the past two decades due to growing precursor emissions, except for some grid cells in the Yangtze River Delta (YRD), the Pearl River Delta (PRD), the Beijing-Tianjin metropolitan area, and other sporadic regions, where MK-Test and Sen's slope identified statistically significant negative O<sub>3</sub> trends. Previous studies have also noted the negative trends over the past decade in these regions (Hu et al., 2024; Li et al., 2022; Wang et al., 2020; Yu et al., 2021). The Beijing-Tianjin area, YRD, and PRD are three critical regions under China's joint efforts to prevent and control air pollution. Regional emission





mitigation in these areas may lead to varying reductions in precursor emissions and declines in O<sub>3</sub> concentrations. Overall, increasing precursor emissions have dominated the O<sub>3</sub> trend in China, as meteorology remained constant during this period. This is evidenced by the linear trends of NOx and NMVOC (Non-methane VOC) emissions in China from 1998 to 2017 (Fig. S9). When comparing the MK-Test and Sen's slope results between the two scenarios model runs, the meteorological conditions from 1998 to 2017 overshadowed the effects of emissions, primarily contributing to the declining O<sub>3</sub> levels (negative trends) in the northernmost regions of China, extending from Inner Mongolia to Northeastern China. Our modeling result shows that mean summer O<sub>3</sub> concentration averaged over Inner Mongolia reached the maximum in 2007 and slightly declined thereafter (Fig. S10). This can be partly attributed to relatively lower precursor emissions resulting from the sparse population and limited industrial activities in this part of China, though having a growing trend (Zhang et al., 2022, 2023).



**Figure 1.** MK-Test and Sen's slope estimated summer  $O_3$  concentration trend and  $O_3$  attribution (fraction or percentage change, %) under baseline (S1) and fixed meteorology (S3) scenarios from 1998 to 2017, respectively. (a) MK-Test and Sen's slope estimated trend from the base scenario S1, the trends of concentrations are well within the MK-Test statistically significant range at 95% confidence level in terms of |Z|>1.96 criteria; (b) same as Fig. 1a but for fixed meteorology scenario S3; (c) mean differences of summer mean  $O_3$  concentrations



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averaged from 1998 to 2017 between the model scenarios 1 and 3, defined by  $\overline{\Delta O}_3 = \overline{O}_{3,S1} - \overline{O}_{3,S3}$ , where  $\overline{O}_{3,S1}$  and  $\overline{O}_{3,S3}$  stand for mean summer O<sub>3</sub> concentrations averaged over 1998 to 2017, respectively.  $\overline{\Delta O}_3$  in three Westernmost provinces of NW is shown by yellow bars and blue bars for three Easternmost provinces of NW. Error bar in Fig. 1c stands for ±1 STD.

To discern potential influences of meteorology on O<sub>3</sub> concentrations at the provincial level, we estimated the differences of summer mean O<sub>3</sub> concentrations averaged from 1998 to 2017 between model scenarios 1 (S1) and 3 (S3), defined by  $\overline{\Delta O}_3 = \overline{O}_{3,S1} - \overline{O}_{3,S3}$ . Figure 1c illustrates  $\overline{\Delta O_3}$  in each province of Northwestern China. Since the mean  $O_3$  levels under scenario S1 were greater than those under scenario S3 with fixed meteorology in 1998, although in some years we also identified negative  $\Delta O_3$  in the three westernmost provinces (Xinjiang, Gansu, and Qinghai), we would expect positive  $\overline{\Delta O}_3$ , as shown in Fig. 1c. In this context, the values of  $\overline{\Delta O}_3$  indicate the extent of meteorological influence on  $O_3$  concentrations. Specifically, the smaller  $\overline{\Delta O}_3$ , the stronger meteorological influences, as a smaller  $\overline{\Delta O}_3$  suggests that  $O_3$  levels with varying meteorology (S1) are closer to those with fixed meteorology in 1998 (S3). We highlighted the three provinces with smaller  $\overline{\Delta O}_3$  (<2 µg m<sup>-3</sup>) using yellow bars, all located in Westernmost NW, indicating a weaker increase in O<sub>3</sub> concentrations in these three westernmost provinces compared to others, aligning well with the RH trends in these provinces, as compared to the other three provinces located in the east of NW (Inner Mongolia, Ningxia, and Shaanxi), as mentioned in the Introduction. As shown in Fig. S11, modeled concentration fractions under the S2 model scenario with fixed emissions in 1998 in the three westernmost provinces of NW are negative, indicating that meteorology tended to inhibit the increase in summer ozone concentrations, particularly in Xinjiang, Gansu, and Qinghai, confirming the result in Fig. 1c. In the other three provinces to the east of NW, we observe larger  $\overline{\Delta O_3}$  values, suggesting that meteorology, along with precursor emissions, tended to enhance O<sub>3</sub> levels from 1998 to 2017. Consequently, we would expect that meteorology favored O<sub>3</sub> growth in these three eastern provinces of NW.

# 3.2 Associations of summer ozone with wetting and warming



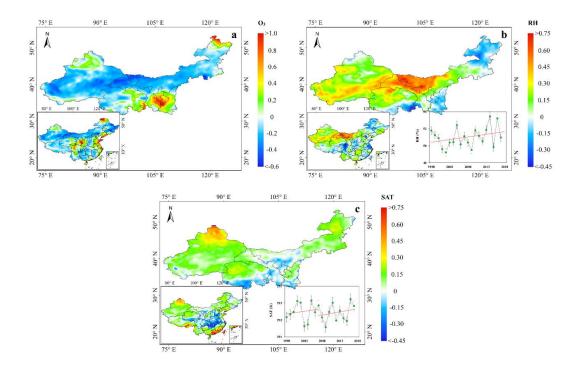


Figure 2. (a) MK-Test statistics for the trends of summer  $O_3$  concentrations from 1998 to 2017 subject to model scenario 2 (S2) with fixed emissions in 1998, (b) MK-Test statistics for summer RH from 1998 to 2017, the line chart on the low-right corner illustrates summer RH in NW from 1998 to 2017 under scenario 2, (c) MK-Test statistics for trend of summer SATs from 1998 to 2017, the line chart shows summer SATs averaged over Northwest China from 1998 to 2017, under scenario 2. Contour figures on the low left corner of Figs. 2a-2c illustrates MK-Test statistics for  $O_3$  trends under fixed emission (model scenario 2) and modeled RH and SAT trends in mainland China from 1998 to 2017. The trends are well within the statistically significant range at 95% confidence level in terms of |Z| > 1.96 criteria.

Figure 2a illustrates the MK-Test statistics of gridded summer O<sub>3</sub> concentrations from 1998 to 2017 subject to the model scenario S2 with fixed emissions and annually varied meteorology, aiming to highlight the effect of meteorology on O<sub>3</sub> evolution. A notable decline in summer O<sub>3</sub> ozone concentrations is evident in most areas of mainland China, except for the Northern China Plain, central China, the Sichuan Basin, and the Guanzhong Plain (inner figure on the left-low corner of Fig. 2a). The declining O<sub>3</sub> trends are also evident in most regions across NW with the most substantial decrease occurring in west Gansu, northwest Inner Mongolia, and Xinjiang. With fixed emission, such an O<sub>3</sub> falling trend is induced primarily by meteorology (climate). Considering that air temperature and humidity are two key meteorological factors influencing O<sub>3</sub> fluctuations, we present the MK-Test statistics of the trends of summer relative humidity (RH, %)

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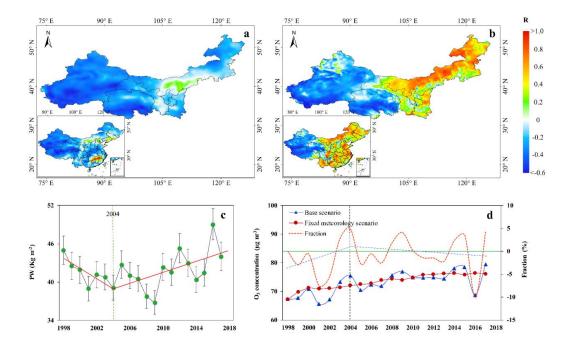
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at the 2 m height in Northwestern and mainland China (illustrated in the lower-left corner of Fig. 2b), along with their annual variations depicted in a line chart in the lower-right corner of Fig. 2b. We also present the surface air temperature (SAT, K) in Northwestern and mainland China in a line chart in the lower-right corner of Fig. 2c. Compared to many places across Eastern and Central China where RH has negative or drying trends (left-low corner of Fig. 2b), RH in NW showed a marked surge, suggesting pronounced wetting, as reported before (Peng and Zhou, 2017). This can also be seen in the annual changes in the mean RH averaged over NW (the line chart on the rightlow corner of Fig. 2b) from 1998 to 2017, dominated by an overall upward RH trend with a mean growth rate of 1.9% per year. Likewise, NW also experienced warming during the same period, as shown in Fig. 2c, particularly in the western part of NW, including Xinjiang, western Gansu Province, and Oinghai Province. NW has been undergoing one of the most significant warming trends in the nation. Between 2005 and 2017, NW, particularly the Xinjiang region, has warmed by over 0.5 K with a rate of 0.04 K/yr. In contrast, eastern and Central China show a cooling trend. The rising SAT trend and falling O<sub>3</sub> trend are in contrast to the common knowledge that increasing SATs should enhance O<sub>3</sub> levels. The result thus implies that SATs did not dominate the O<sub>3</sub> trend in NW.

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**Figure 3.** (a) Pearson correlation (R) between summer  $O_3$  concentration and RH from 1998 to 2017 under S2 scenario, (b) R between summer OH radical concentration and  $O_3$  concentration from 1998 to 2017 under S2 scenario, (c) summer surface precipitable water (PW) in the bounded area of NW from 1998 to 2017, error bar stands for  $\pm 1$  STD and red solid line stands for linear trend, (d) summer  $O_3$  concentrations under Scenarios 1 (dark blue solid line with triangles) and 3 (red solid line with circles) scaled on the left-Y axis, and their fractions  $F_{O3} = (C_{S1} - C_{S3}) / C_{S3} \times 100\%$  (red dashed line) in three Westernmost provinces of HW (Xinjiang, Qinghai, and Gansu), where  $C_{S1}$  and  $C_{S3}$  are the summer (June to August) surface  $O_3$  air concentrations from the base (S1) and fixed meteorology (S3) scenarios, scaled on the right-Y axis, the blue dashed line in Fig. 3d is a linear fitting line from 1998 to 2004 and 2004 to 2017, respectively. The green solid line represents the zero-value line of the fraction.

As mentioned earlier, since rising temperatures favor O<sub>3</sub> formation while increasing humidity is detrimental to O<sub>3</sub> growth, it is intriguing to investigate which of these two meteorological factors contributes more to the O<sub>3</sub> trend in NW. We first calculated linear correlation coefficients between RH and O<sub>3</sub> concentrations modeled from the S2 scenario (fixed emissions) from 1998 to 2017 across Northwestern and mainland China (bottom left corner of Fig. 3a). Strong negative correlations are evident in most regions of Xinjiang, Qinghai, and Western Gansu. These negative correlations indicate declining O<sub>3</sub> concentrations with rising RH. To highlight the influences of RH and precipitation on summer O<sub>3</sub> variation, Fig. 3d illustrates modeled mean summer O<sub>3</sub> concentrations averaged over three Westernmost provinces of HW (Xinjiang, Qinghai, and Gansu)



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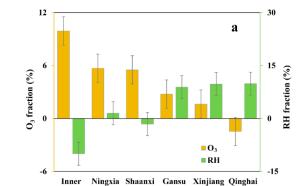
under the base scenario S1 with annually varying summer meteorology and emissions, and S3 with fixed meteorology in 1998 and annually varying emissions averaged over NW, along with the concentration fractions (%, red dashed line) between the two scenarios from 1998 to 2017. O<sub>3</sub> concentrations exhibit the same increasing trend primarily driven by rising precursor emissions. However, the S1 scenario shows more significant annual fluctuations (solid dark blue line in Fig. 3d) compared to the S3 scenario (solid red line in Fig. 3d). These stronger fluctuations can be attributed to the influence of meteorology. Since scenarios 1 and 3 shared the same emissions and chemistry, the concentration fractions quantify the contributions of meteorology to the O<sub>3</sub> trend alone. As shown, the fractions increased from 1998 to 2004, decreased thereafter, and turned negative since 2009 (the red dashed lines), indicating that meteorology has led to declining O<sub>3</sub> concentrations since 2005 in NW. Among the two key meteorological factors influencing O<sub>3</sub> formation, increasing SAT (Fig. 2c) tends to elevate O<sub>3</sub> levels, which contradicts the decreasing and negative O<sub>3</sub> fractions. Consequently, RH likely plays a more significant role in the O<sub>3</sub> trend in NW, as rising RH aligns well with the decreasing and negative O<sub>3</sub> fractions.

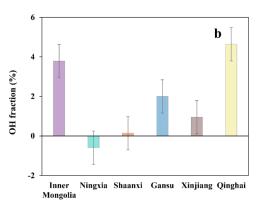
Figure 3c displays the mean summer surface precipitable water (PW, kg/m2) over the region bounded between 75 – 100°E in Northwest China from 1998 to 2017, collected from the NCAR reanalysis (https://psl.noaa.gov/cgi-bin/data/composites/printpage.pl). One can identify a slow decrease in PW from 1998 to 2004 and a rapidly growing PW thereafter. Since warming exacerbates drought in arid and sub-arid NW (Lian et al., 2021), growing precipitation in NW implies that increasing humidity might play a more significant role than warming in the changing climate, which acts to reduce O<sub>3</sub> levels via precipitation washout and chemistry. A close look at Figs. 2b, 2c, and Fig. 3a indicate that more substantial warming and wetting occurred in Xinjiang, Gansu (including westernmost of Inner Mongolia), and Qinghai provinces. We compared the fractions of provincial mean summer RH and summer O<sub>3</sub> concentrations from the fixed emissions and annually varying meteorology scenario (S2) between 1998 and 2017 (Fig. 4a), defined by f =  $(Y_{2017} - Y_{1998}) / Y_{1998} \times 100\%$ , where f is the concentration or RH fractions and  $Y_{1998}$  and  $Y_{2017}$  stand for O<sub>3</sub> concentrations or RH in 1998 and 2017, respectively, thereby to highlight the net effect from meteorology. The most significant fraction of O<sub>3</sub> concentration was identified in Inner Mongolia at 9.9%, followed by Ningxia (5.7%), Shaanxi (5.5%), Gansu (2.8%), Xinjiang (1.6%), and Oinghai (-1.5%), respectively. Among the six provinces, Xinjiang and Gansu accounted for smaller O<sub>3</sub> fractions, and Qinghai accounted for a negative fraction (Fig. 4a), indicating that O<sub>3</sub>





levels in these three provinces inclined less significantly or even declined (Qinghai) between 1998 and 2017 than the other three provinces. Interestingly, more significant O<sub>3</sub> fractions, meaning growing O<sub>3</sub> levels for the two decades in Inner Mongolia, Ningxia, and Shaanxi, correspond to smaller (Ningxia) or negative (Inner Mongolia and Shaanxi) RH fractions, meaning falling RH. In contrast, more diminutive and negative O<sub>3</sub> fractions in Xinjiang, Gansu, and Qinghai provinces are associated well with the stronger increasing trend of summer RH by MK-test (Figs. 2b and 4a) at the growing rate of 9.8% in Xinjiang, 8.9% in Gansu, and 9.9% in Qinghai from 1998 to 2017, respectively. While the weak or falling summer O<sub>3</sub> levels in three provinces respond to both growing RH and SAT trends (Fig. 2b and 2c), since growing SATs tend to enhance O<sub>3</sub> concentrations but modeled O<sub>3</sub> respond weakly to rising SAT in these three provinces, the result demonstrates again that the wetting might inhibit O<sub>3</sub> formation and increment for the past decades in NW.





Mongolia

**Figure 4.** Summer RH,  $O_3$ , and OH radical fractions between 1998 and 2017 in six provinces of NW under the fixed emission scenario (S2). (a) RH and  $O_3$  fractions,  $O_3$  fractions are scaled on the left Y-axis, and RH fractions are scaled on the right Y-axis, (b) OH radical fractions. Error bars denote  $\pm 1$  STD.

It is worthwhile to point out that model scenario 2 with fixed emissions also considers chemistry, in which OH radicals and water vapor can interact. The primary source of OH radicals in the atmosphere is the reaction between water vapor  $(H_2O)$  and excited oxygen atoms  $(O(^1D))$ , which are formed by the photolysis of ozone  $(O_3)$  by ultraviolet (UV) radiation. The OH radicals react with ozone to form oxygen  $(O_2)$  and the excited oxygen atom  $O(^1D)$ , which can further react with water vapor to regenerate two OH radicals:



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$$O_3 + OH \rightarrow O_2 + O(^1D),$$
  
351  $O(^1D) + H_2O \rightarrow 2 OH.$  (2)

This reaction does not lead to a net reduction in OH radicals but contributes to ozone removal by initiating reactions that lead to the formation of less reactive species. Figure 3b is a correlation diagram between modeled summer O3 concentrations and OH radicals in NW and mainland China (the left-low corner of Fig. 3b) subject to the fixed emission scenario (S2). The spatial pattern of correlation coefficients in NW is, to some extent, similar to that between O3 concentrations and RH (Fig. 3a), showing the negative associations between OH radicals and O<sub>3</sub> concentrations, particularly in Xinjiang and Oinghai with the most significant wetting trend, suggesting that increasing OH radicals reduce O<sub>3</sub> levels. Figure 4b displays the fractions of provincial mean summer OH radicals from the fixed emissions and annually varying meteorology scenario (S2) between 1998 and 2017. Relatively larger positive fractions can be discerned in Xinjiang, Qinghai, and Gansu, meaning increasing OH radicals in these provinces as a result of increasing RH (Eq. 1), which corresponds to higher RH fractions and lower O<sub>3</sub> fractions. An exception occurs in Inner Mongolia, where the large positive OH radical fraction (Fig. 4b) is associated with negative RH and large O<sub>3</sub> fractions (Fig. 4a). OH radicals show an increasing trend (Fig. S12) which agrees with the positive correlation between OH and O<sub>3</sub> concentrations shown in Fig. 3b. This puzzling from the modeling results needs further investigations.

There are several primary mechanisms of OH radicals leading to  $O_3$  reduction, including the reaction with VOCs to form peroxyl radicals (RO<sub>2</sub>), the reaction of RO<sub>2</sub> with nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) to form nitrogen dioxide (NO<sub>2</sub>) and other compounds, and photodissociation of NO<sub>2</sub>. Figures S9a and S9b show the MK test estimated NO<sub>x</sub> and NMVOC emission trends from 1998 to 2017. While increasing NO<sub>x</sub> is observed in NW (Fig. S9a), its emission levels are considerably lower than in Eastern and Southern China (Fig. S9c). Both NMVOC emission levels and trends in NW are significantly smaller than in Eastern and Southern China due to lower populations and less developed economies (Fig. S9b and S9d). Hence, it is likely that the reaction of water vapor (H<sub>2</sub>O) with excited oxygen atoms (O( $^1$ D)) produced by the photodissociation of ozone by UV light plays a specific role in the negative correlation between O<sub>3</sub> and OH radicals.

A multiple regression model (MRM) was established to further quantitatively assess the contribution of RH and SAT to summer O<sub>3</sub> concentrations in NW. The MRM has been applied in





a number of investigations to quantify meteorological effects on ozone evolution in China (Li et al., 2019, 2020, 2021; Yan et al., 2024). For SAT and RH, the MRM model can be defined as

$$\ln (O_3) = \alpha + \beta_1 \ln (SAT) + \beta_2 \ln (RH) + \varepsilon, \tag{3}$$

where  $\alpha$ ,  $\beta_1$ , and  $\beta_2$  are model coefficients, and  $\varepsilon$  is a random error, respectively. In the first instance, we considered three provinces with significant wetting trends: Xinjiang, Gansu, and Qinghai. We combined annual summer O<sub>3</sub>, RH, and SAT from three provinces into one time series of each dependent and independent variable from 1998 to 2017, yielding 60 samples for each variable. Tables S4-S6 present regression coefficients and statistics of the MRM. To examine the multicollinearity in the MRM, we estimated the variance inflation factor (VIF, Table S4-S6). The VIFs for RH and SAT are far less than 10, indicating no multicollinearity in the model. The contribution of RH and SAT to O<sub>3</sub> is calculated by:

391 Contribution<sub>i</sub> = 
$$\frac{\left|\beta_{i} \times \frac{STD_{i}}{STD\alpha}\right|}{SUM}$$
, (4)

where SUM =  $\sum_i |\beta_i \times STD_i/STD_{\alpha}|$ , and  $\beta_i$  denotes the regression coefficient of independent variable i (namely, RH and SAT),  $STD_i$  is the standard deviation of regression coefficient of independent variable i, and  $STD_{\alpha}$  represents the standard deviation of coefficient of intercept.

The MRM analysis reveals that RH contributes 77.5%, 59.4%, and 46.2% to O<sub>3</sub> variations in Xinjiang, Qinghai, and Gansu. Accordingly, SAT contributes 22.5%, 40.6%, and 53.8% to O<sub>3</sub> fluctuations in these three provinces from 1998 to 2017. The results indicate that RH has become a major contributor to long-term change in O<sub>3</sub> concentrations, overwhelming or equivalent to SAT's contributions to the O<sub>3</sub> trend for the past two decades, likely via interactions among increasing precipitation washout and OH radical, O<sub>3</sub> concentration, and water vapor.

We further assess the contributions of precipitation, RH, SAT, winds, and PBLH (planetary boundary-layer height) to O<sub>3</sub> evolution in NW using an extended MRM model. The results confirm that RH and SAT made the most significant contributions to long-term ozone trends than other meteorological variables under the long-term perspective. As seen from Fig. 5, the contribution of RH (49.5%) and SAT (47.6%) to O<sub>3</sub> evolution in NW was almost equivalent (Fig. 5a), but the SAT (92.4%) dominated in entire mainland China (Fig. 5b). Whereas the PBLH only contributed



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about 2% to  $O_3$  variations because the PBLH varies on a daily and hourly basis and affects insignificantly (summer) seasonal changes in  $O_3$  concentrations.

b a 47.59% Precipitation:0.21% 92.37% Wind:0.26% PBLH RH 2.42% SAT Northwest China China Precipitation ■ Wind 1.85% 0.63% Wind:0.03% 49.51%

**Figure 5.** Contribution of PBLH (planetary boundary-layer height), RH, SAT, precipitation, and wind to  $O_3$  evolution. (a) Northwest China, (b). Mainland China.

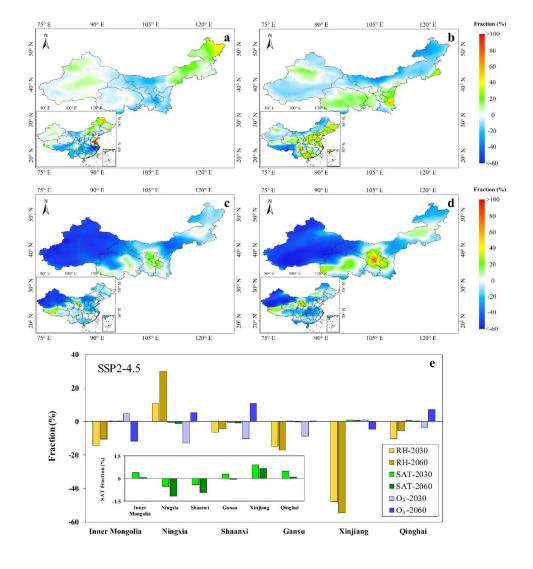
It should be pointed out that since the wetness also helps growing vegetation, which causes increasing biogenic VOC emission (BVOC) and hence rising O<sub>3</sub> concentrations, the contribution of BVOCs to O<sub>3</sub> fluctuations under increasing wetness should also be taken into consideration. To examine this hypothesis, we designed an extra model scenario (S5) in which we used the updated LUC data (section 2.4) and fixed LUC in 1998 in the WRF-Chem simulation and ran the model from 1998 to 2017. Since the LUC defines vegetation coverage, the fixed LUC suggests no vegetation change during this period. We then estimated the fractions between modeled O<sub>3</sub> concentration with fixed LUC and the baseline scenario S1 with annually-varying LUC, thereby eliminating the effects from emissions, meteorology, and chemistry but highlighting the LUC (or greenness) effect only. The result is illustrated in Fig. S13. As shown, while the fraction fluctuates positively (Xinjiang) and negatively (Northern Shaanxi and Eastern Inner Mongolia), indicating increasing and decreasing O<sub>3</sub> levels induced by LUC or greening, the magnitudes of provincial mean concentration fractions are very small, mostly ranging from -1% to 0.3%, suggesting that the LUC under the wetting trend in NW plays an insignificant role in O<sub>3</sub> fluctuation.

# 3.3 Projected future climate impacts on summer Ozone





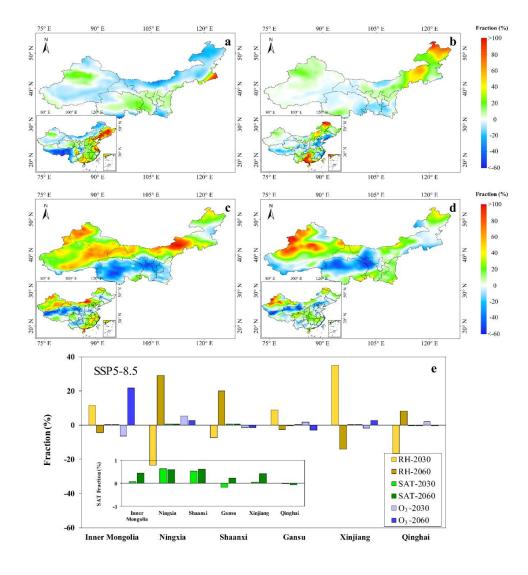
Given the significant impacts of wetting on O<sub>3</sub> evolution in NW over the past two decades, concerns have also been raised regarding O<sub>3</sub> responses to future wetting and warming. We estimated the projected O<sub>3</sub> concentrations, RH, and SAT fractions between the baseline and fixed meteorology scenarios under the SSP2-4.5 and SSP5-8.5 pathways (section 2.6) for the years 2019, 2030, and 2060, respectively (Figs. 6 and 7). As mentioned earlier, since both scenarios implemented the same emissions and chemistry but the baseline scenario accounted for variable meteorology, the fractions (or differences) between the two scenarios eliminated the effects of emissions and chemistry, thereby isolating meteorological signals only.







**Figure 6.** (a) Summer O<sub>3</sub> concentration fractions of 2019 to 2030 under SSP2-4.5scenario, estimated by  $F_{\text{SSP245}} = (C_{2030} - C_{2019}) / C_{2019} \times 100\%$ , where  $C_{2019}$  and  $C_{2030}$  are the summer surface O<sub>3</sub> air concentrations in 2019 and 2030, respectively, (b) same as Fig. 6a but for O<sub>3</sub> fractions of 2019 to 2060, (c) same as Fig. 6a but for summer RH fractions of 2019 to 2030, (d) same as Fig. 6a but for summer RH fractions of 2019 to 2060, (e) fractions of RH, SAT, and O<sub>3</sub> of 2019 to 2030 and 2019 to 2060 (shown in the same color bar as 2030 but with oblique lines) in the six provinces of Northwestern China under the SSP2-4.5 scenario.



**Figure 7.** (a) Summer O<sub>3</sub> concentration fraction of 2019 to 2030 under the SSP5-8.5 scenario, estimated by  $F_{\text{SSP585}} = (C_{2030} - C_{2019}) / C_{2019} \times 100\%$ , where  $C_{2019}$  and  $C_{2030}$  are the summer surface O<sub>3</sub> air concentrations in 2019 and 2030, respectively, (b) same as Fig. 7a but for O<sub>3</sub> fraction of 2019 to 2060, (c) same as Fig. 7a but for summer RH fraction of 2019 to 2030, (d) same as Fig. 7a but for summer RH fraction of 2019 to 2060, (e)



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fractions of RH, SAT, and  $O_3$  of 2019 to 2030 and 2019 to 2060 (shown in the same color bar as 2030 but with oblique lines) in the six provinces of Northwestern China under the SSP5-8.5 scenario.

Detailed discussions on the associations between projected O<sub>3</sub> evolution and trends of RH and SAT under the SSP2-4.5 and SSP5-8.5 pathways from 2019 to 2030 and from 2019 to 2060 are presented in Section S4 in the Supplement and Figs. 6-7 and Fig. S14, respectively. Overall, from 2019 to 2030, we observe a stronger negative association between O<sub>3</sub> and RH under SSP5-8.5 than under SSP2-4.5 across NW, suggesting that climate conditions under the SSP5-8.5 scenario will more effectively suppress O<sub>3</sub> growth compared to SSP2-4.5 during this period. However, from 2019 to 2060, the SSP5-8.5 tends to project higher O₃ levels than SSP2-4.5. From a wetting trend perspective, we must determine which pathway would project a more realistic future humidity trend in NW. China has been making significant strides in transitioning to renewable energy sources, reducing its carbon footprint, and committing to achieve a carbon peak before 2030 and carbon neutrality by 2060. These commitments align more closely with the mitigation efforts outlined in SSP2-4.5, which, as manual interventions, will overshadow the impact of climate change on O<sub>3</sub> fluctuations. There have also been debates regarding ongoing wetting trends in NW based on CMIP6 projections under SSP5-8.5 and historical data analysis (Huang et al., 2017). Historically, China's rapid economic growth has heavily depended on fossil fuels, which may render SSP5-8.5 more applicable in China. The responses of O<sub>3</sub> evolution to RH under SSP5-8.5 are more akin to those observed from 1998 to 2017 (section 3.2) than to SSP2-4.5. In light of this, while China may achieve its carbon peak and neutrality goals, the wetting trend in NW will likely persist in the coming decades, exacerbating continuous O<sub>3</sub> pollution in NW, in addition to manual interventions.

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## 4. Conclusions

This study conducted extensive sensitivity model simulations to explore the surface O<sub>3</sub> responses to past and future climate change in Northwestern China. The results demonstrated that significant wetting since the 21st century in this region, particularly in the three westernmost provinces (Xinjiang, Qinghai, and Gansu) of NW, has slowed O<sub>3</sub> growth. We show that in these provinces, the wetting influence on O<sub>3</sub> evolution has outweighed the warming effect and played an almost equal role to warming across the entire NW. Both factors contributed approximately 50%





479 changes in O<sub>3</sub> concentrations. This finding contrasts with the dominant role of warming in the 480 meteorological effects on O<sub>3</sub> evolution in China. We linked the increasing OH radicals, which 481 interact with water vapor, to the declining O<sub>3</sub> trend. Given that humidification in NW is projected 482 to continue in the coming years and that the mitigation of O<sub>3</sub> precursor emissions in China will 483 still be implemented under the nation's air pollution control strategies, the impact of 484 humidification on O<sub>3</sub> trends in NW will likely become more significant. Such an impact should be 485 considered in future policymaking for the reduction of O<sub>3</sub> pollution in NW. 486 487 Data availability 488 **ERA-Interim** reanalysis data be accessed can 489 https://www.ecmwf.int/en/forecasts/datasets/reanalysisdatasets/era-interim. EDGAR dataset is 490 available at https://edgar.jrc.ec.europa.eu/. LUC data can be accessed at http://www.resdc.cn. 491 DPEC Inventory is available at http://meicmodel.org.cn. The meteorological data under the 492 CMIP6 framework is collected from https://aims2.llnl.gov/search/cmip6/. 493

to summer O<sub>3</sub> variations from 1998 to 2017, among the major meteorological factors causing

# 494 **Author contributions**

- 495 XZ and YY contribute equally to this article. All authors contributed to the manuscript and have given approval of the final version. XZ and HG coordinated and supervised the project. XZ, YY,
- 497 WW and JM designed the present experiment, carried out modeling, and drafted the manuscript.
- 498 NZ, HS, XJ, CW, and HM collected the data. XZ, YY, ZY and TH analyzed simulation results.

### 500 Competing interests

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The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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