1	Impacts of compound extreme weather events on ozone in the present and future
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- 39 Abstract
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41 The Weather Research and Forecasting model with Chemistry (WRF/Chem) was 42 used to study the effect of extreme weather events on ozone in the US for historical 43 (2001-2010) and future (2046-2055) periods under the RCP 8.5 scenario. During 44 extreme weather events, including heat waves, atmospheric stagnation, and their 45 compound events, ozone concentration is much higher compared to non-extreme events 46 period. A striking enhancement of effect during compound events is revealed when heat 47 wave and stagnation occur simultaneously as both high temperature and low wind speed 48 promote the production of high ozone concentrations. In regions with high emissions, 49 compound extreme events can shift the high-end tails of the probability density 50 functions (PDFs) of ozone to even higher values to generate extreme ozone episodes. 51 In regions with low emissions, extreme events can still increase high ozone frequency 52 but the high-end tails of the PDFs are constrained by the low emissions. Despite large 53 anthropogenic emission reduction projected for the future, compound events increase 54 ozone more than the single events by 10% to 13%, comparable to the present, and high 55 ozone episodes with maximum daily 8h average (MDA8) ozone concentration over 56 70ppbv are not eliminated. Using the CMIP5 multi-model ensemble, the frequency of 57 compound events is found to increase more dominantly compared to the increased 58 frequency of single events in the future over the US, Europe, and China. High ozone 59 episodes will likely continue in the future due to increases in both frequency and 60 intensity of extreme events, despite reductions in anthropogenic emissions of its 61 precursors. However, the latter could reduce or eliminate extreme ozone episodes, so 62 improving projections of compound events and their impacts on extreme ozone may 63 better constrain future projections of extreme ozone episodes that have detrimental 64 effects on human health.

Key words: WRF/Chem, heat waves, stagnation, compound event, high surface ozone

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70 1. Introduction

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72 Tropospheric ozone is a secondary air pollutant resulting from complicated 73 photochemical reactions in the presence of its precursors such as volatile organic 74 compounds, NO_x, and CO (Placet et al., 2000). During the past decades, ozone pollution 75 has been of increasing concern to the public because excessive ozone may have an 76 adverse effect on human health such as increased risk of death (Filleul et al., 2006). 77 Ozone also has important effects on agriculture, constructions, and ecology (Weschler, 78 2006; Gryparis et al., 2004). Moreover, as a greenhouse gas, increasing concentrations 79 of ozone may amplify global warming (Mitchell, 1989; Schimel et al., 2000). Thus, it 80 is important to understand factors that govern ozone concentration in a perturbed 81 environment.

82 Ozone formation is particularly active when favorable meteorological conditions 83 coincide with the presence of high precursor emissions (Sharma et al., 2017; Agrawal 84 et al., 2003). Meteorological factors that are closely related to ozone formation include 85 daily maximum temperature (Fiore et al., 2015), wind speed, cloud cover (Jacob and Winner, 2009; Otero et al., 2016), etc. Using dynamical downscaling to develop high 86 87 resolution climate scenarios, Souri et al. (2016) found significant ozone increase in the 88 US during heat wave events, with regional mean maximum daily 8 h average (MDA8) 89 O₃ increases roughly by 0.3 ppbv to 2.0 ppbv compared with non-heat wave period 90 under RCP 8.5. Based on observed data in the US from 2001-2010, Flynn et al. (2010) 91 found significant ozone increase during heat waves in particular for high ozone concentration (i.e., 95th percentile ozone increased by 25%) and PM_{2.5} increase during 92 atmospheric stagnation (i.e., 95th percentile ozone increased by 65%). Both heat waves 93

94 (Gao et al., 2013; Hou and Wu, 2016; Gao et al., 2012) and atmospheric stagnation
95 (Sillmann et al., 2013) have been projected to increase substantially in the future,
96 suggesting significant impacts on ozone and PM_{2.5} in the future.

97 Going beyond traditional study of single extreme weather events and their impacts, 98 compound effect of extreme events has been explored in recent studies (Meehl and 99 Tebaldi, 2004). Compound effect can be defined using different criteria including: 1) 100 two or more extreme events occurring simultaneously or successively; 2) combinations 101 of extreme events potentially reinforcing each other; 3) two or more events combined 102 to become an extreme event even though the events themselves are not extreme (Horton 103 et al., 2014; Zscheischler and Seneviratne, 2017). The compound effect of more than 104 one extreme weather event has been shown to potentially have a higher impact than a 105 single extreme weather event alone. For example, Leonard et al. (2014) concluded that 106 compound effect could be higher than simple additive effect. As an example, they found 107 that the compound effect of heat waves and drought on the global carbon cycle exceeds 108 the additive effect of the individual events. For ozone, heat waves and atmospheric 109 stagnation are two key environmental factors that may lead to compound effect, as high 110 surface temperature under atmospheric stagnation with low wind speed, clear sky, and 111 reduced precipitation and soil moisture may escalate into a heat wave. This motivates 112 the present study to investigate the compound effect of simultaneous occurrence of heat 113 waves and atmospheric stagnation on ozone pollution.

114 Model output from the Coupled Model Intercomparison Project phase 5 (CMIP5; 115 Seneviratne et al. (2012)) has been widely used to investigate climate change and its 116 impacts. Using a multi-model ensemble such as CMIP5 is particularly important for 117 studying high-impact and low-probability extreme events to yield more robust analyses 118 (Zscheischler et al., 2014; Taylor et al., 2012; Sillmann et al., 2013). However, air 119 quality is significantly influenced by regional processes such as cloudiness and mesoscale circulation as well as local emissions. With high spatial and temporal 120 121 resolutions and more detailed representations of chemical reactions and emission inventory (Diffenbaugh and Giorgi, 2012), regional climate and chemistry models are
useful tools that have been widely adopted to study air quality and impact of climate
change on air quality (Kharin et al., 2013; 2013; Gao et al., 2012; Leung and Gustafson,
2005; 2010). This study combines analysis of regional online-coupled meteorologychemistry simulations and analysis of the CMIP5 multi-model ensemble to investigate
the impact of extreme weather events on ozone concentration in the present and future
climate.

In what follows, we first investigate the ability of the regional climate-chemistry model in reproducing the observed extreme weather events and ozone concentration in the US. Following the evaluation, the impact of single and compound extreme weather events on ozone concentration at present and future is examined. Lastly, future changes of extreme weather events are discussed in the broader context of the multi-model CMIP5 ensemble.

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136 **2. Model description and configuration**

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138 In this study, a modified version of WRF/Chem v3.6.1 (Yahya et al., 2017a) was adopted for regional simulations. The detailed modification has been described in 139 140 Yahya et al. (2017b), but the main new features include the extended Carbon Bond 2005 141 (CB05) of Yahya et al. (2016) gas-phase mechanism with chlorine chemistry of Yahya 142 et al. (2016). The anthropogenic emissions used in WRF/Chem were based on the 143 emissions in RCP8.5 (Yarwood et al., 2005; Sarwar and Bhave, 2007) and detailed 144 information of processing the RCP 8.5 emission to model-ready format is available in Moss et al. (2010). Biogenic emissions were calculated online in WRF/Chem 145 146 depending on the meteorology at present or future using the Model of Emissions of 147 Gases and Aerosols from Nature version 2 (van Vuuren et al., 2011). The 148 meteorological and chemical initial and boundary conditions for WRF/Chem were downscaled from simulations provided by the modified CESM/CAM version 5.3 149

150 (referred to as CESM NCSU) (Yahya et al., 2017b; Guenther et al., 2006; 2014; He 151 and Zhang, 2014). Glotfelty et al. (2017) documented the details of the downscaling method and provided a comparison of some meteorological parameters simulated by 152 153 CESM NCSU and CESM in CMIP5, showing consistent performance between the two 154 CESM versions. Two simulation periods using WRF/Chem were selected in this study: 155 a historical period (2001-2010) and a future period (2046-2055), and simulations were 156 performed over the contiguous US (Fig. 1), with a horizontal grid spacing of 36 km and 157 34 vertical layers from surface to 100 hPa. The simulations for the historical period 158 have been comprehensively evaluated against surface and satellite observations in 159 Yahya et al. (2017a) and the projected changes in climate, air quality, and their 160 interactions for the future period have been analyzed in Yahya et al. (2017b). However, 161 those results have not been previously evaluated for climate extremes and their impacts 162 on surface O₃, which is the focus of this work.

163 In addition to the regional model results, output from the CMIP5 (https://esgfnode.llnl.gov/search/cmip5/) multi-model ensemble was used in this study to elucidate 164 165 the impact of climate change on compound extreme weather events. A total of 20 166 CMIP5 models were selected in this study, and the list of models is shown in Table 1. 167 Variables used in this study mainly include daily maximum near-surface air temperature, 168 daily precipitation, daily mean near-surface wind speed and daily mean 500 hPa wind speed, and the data were interpolated to a spatial resolution of $2^{\circ} \times 2^{\circ}$. Three periods 169 170 were selected with two periods that overlap in part with that of the regional simulations 171 (1991-2010 as historical period and 2041-2060 in RCP 8.5), and an additional period 172 extending to the end of this century (2081-2100).

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Table 1 A list of the CMIP5 models used in this study

Reference

Glotfelty and

Zhang (2016)

Yahya et al.

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Arora et al.

Scoccimarro et al. (2011)

Weare et al.

al.

al.

al.

et

(2017b)

(2013) Dix

(2013) Xin

(2012)

(2011)

(2012)Rotstayn

(2011)

(2011)

(2010)

Dufresne

al. (2013)

et

al. (2010) Donner et al.

Jones et al.

Volodin et al.

Bi

Model	Institution	Resolution (Lon×Lat)
1. ACCESS1.0	Commonwealth Scientific and Industrial Research Organization	1.875×1.25
2. ACCESS1.3	(CSIRO), Australia and Bureau of Meteorology (BOM), Australia	1.875×1.25
3. BCC-CSM1.1	Beijing Climate Center, China Meteorological Administration	2.81×2.77
4. CanESM2	Canadian Centre for Climate Modeling and Analysis, Canada	2.81×2.79
5. CMCC-CM	Euro-Mediterraneo sui	0.75×0.75
6. CMCC-CMS	Cambiamenti Climatici, Italy	1.875×1.86
7. CSIRO_Mk3.6.0	Commonwealth Scientific and Industrial Research Organization (CSIRO), Australia	1.875×1.86
8. GFDL-ESM2M	NOAA Geophysical Fluid	2.5×2.0
9. GFDL-ESM2G	Dynamics Laboratory, USA	2.5×2.0
10. HadGEM2_CC	Met Office Hadley Centre, UK	1.875×1.25
11. INM-CM4	Institute for Numerical Mathematics, Russia	2.0×1.5
12. IPSL-CM5A- LR		3.75×1.875
13. IPSL-CM5A- MR	Institut Pierre-Simon Laplace, France	2.5×1.25
14. IPSL-CM5B- LR		3.75×1.875
15. MIROC-ESM	Atmosphere and Ocean Research Institute (The University of	2.81×1.77
16. MIROC-ESM-	Tokyo), National Institute for	2.81×1.77

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CHEM

17. MIROC5

18. MPI-ESM-LR

19. MPI-ESM-MR

Marine-Earth

2.81×1.77

Environmental Studies and Japan

for

Agency

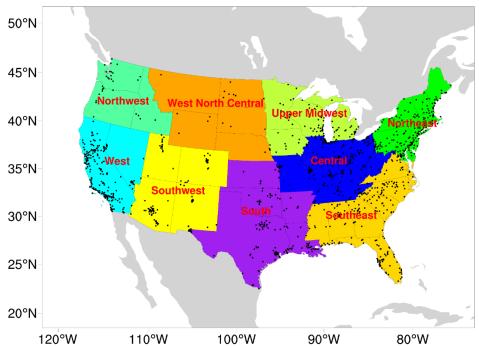
Max

20. MRI-CGCM3	Meteorological	Research	1.125×1.125	Watanabe	et
20. IVIKI-CGCIVI3	Institute, Japan		1.123^1.123	al. (2010)	

182 **3. Evaluation of meteorology and ozone**

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The System (downloaded 184 Air Quality (AQS) dataset from 185 https://www.epa.gov/aqs) was used in this study to evaluate how well the WRF/Chem model performs in simulating ozone concentrations, particularly high ozone 186 187 concentrations that are more strongly related to extreme weather events. The locations 188 of observation stations in AQS are shown in Fig. 1 and overlaid on nine climate regions 189 in the US (Karl and Koss, 1984). For evaluation of simulated extreme weather events, 190 the NCEP North American Regional Reanalysis (Zanchettin et al., 2013) dataset was 191 used. 192



193120°W110°W100°W90°W80°W194Fig. 1. The WRF/Chem simulation domain and climate regions in the US. The red195points (~ 1200) represent the observation stations of O3 in AQS.

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197 **3.1 Evaluation of extreme weather events**

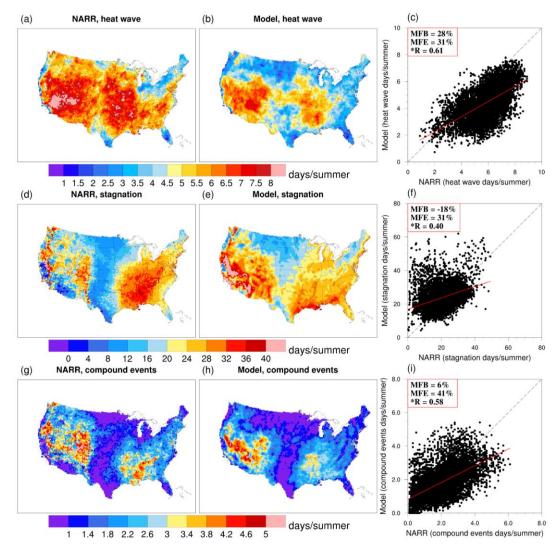
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199 Two types of extreme weather events including heat waves and atmospheric

200 stagnation, as well as their compound events were investigated, considering their close 201 relationship with ozone pollution (Hou and Wu, 2016). A heat wave is defined to occur 202 when daily maximum 2-meter air temperature exceeds a certain threshold continuously 203 for three days or more. The threshold is set as the 97.5th percentile of the historical 204 period (2001-2010 for WRF/Chem and 1991-2010 for CMIP5 in this study) and is 205 location dependent to take into account the wide-ranging characteristics of different 206 regions (Yukimoto et al., 2012; Mesinger et al., 2005). An atmospheric stagnation day 207 is defined to occur when daily mean 10-m wind speed, daily mean 500 hPa wind speed, 208 and daily total precipitation are less than 20% of the climatological mean condition 209 (2001-2010 for WRF/Chem in this study) (Gao et al., 2012; Meehl and Tebaldi, 2004). 210 A compound event occurs when both heat wave and atmospheric stagnation occur 211 simultaneously on the same day. For each grid, the same threshold determined for the 212 present period is used for the future period to evaluate the future changes.

213 To evaluate the ability of the regional model in reproducing the extreme weather 214 events, Fig. 2 shows the distribution of mean number of summer heat wave days, 215 atmospheric stagnation days, and compound event days corresponding to coincidental 216 heat wave and atmospheric stagnation during 2001-2010. Observations based on the 217 NARR dataset and the model results are shown, along with scatterplots comparing the 218 observations and simulations at each NARR grid point over land. Statistical metrics, including mean fractional bias (MFB), mean fractional error (MFE) and correlation 219 220 coefficient (R), based on the formulae (A2), (A3) and (A6) in the appendix, are shown 221 in the scatterplots.

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225 Fig. 2. Distribution of mean number of extreme weather days in summer of 2001-2010 from observations (NARR; left panels) and model simulations (middle panels) 226 and scatterplots comparing them at each NARR grid point over land (right panels) for 227 228 heat wave days (Figs. 2a,b,c), atmospheric stagnation days (Figs. 2d,e,f) and 229 compound event days (Figs. 2g,h,i). The numbers located on the top left of the 230 scatterplots (Fig. 2c,f,i) indicate the statistical metrics including mean fractional bias 231 (MFB), mean fractional error (MFE) and correlation coefficient (R). A r-test (a=0.05) 232 for the linear correlation coefficient was performed and *R indicates statistical 233 significance at 95% confidence level. The red solid lines in the scatterplots are the 234 linear regression lines, and the black dashed lines are one-to-one reference lines. 235

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The spatial distributions of both heat waves and atmospheric stagnation are generally consistent between NARR and WRF/Chem (top and middle rows). For example, for heat waves (Figs. 2a,b), the model captures the high frequency of occurrence in the western US and eastern central US albeit widespread 240 underestimations particularly in the northern US and the central Great Plains. For 241 atmospheric stagnation (Figs. 2d,e), the observed dipole feature of high frequency of 242 occurrence in the western and eastern US, separated by the central Great Plains, is well 243 reproduced by the model but biases in the magnitude are noticeable. To quantitatively 244 evaluate the simulations, the WRF/Chem model results were bilinearly interpolated to the NARR grid suggested by Horton et al. (2014), and scatterplots were drawn to show 245 246 the results for all the NARR grid points (Figs. 2c,f). No benchmark is available regarding the statistical metrics for extreme weather events but we adopt the 247 248 benchmarks widely used in air quality studies. For example, Hou and Wu (2016) 249 suggested 15%/35% (MFB/MFE) for O₃ and 50%/75% (MFB/MFE) for PM2.5 species. 250 From this perspective, the MFB and MFE for either heat waves or atmospheric 251 stagnation are within or close to the benchmarks for O_3 , and well within the benchmarks 252 for PM_{2.5} species. Moreover, the model results are correlated with NARR, with R equals 253 to 0.61 and 0.40, respectively, for heat waves and atmospheric stagnation and 254 statistically significant at 95% confidence level.

255 The western US receives most of its precipitation in the cold season when the 256 North Pacific jet stream steers storm tracks across the region (Neelin et al., 2013). 257 During summer, the North Pacific subtropical high pressure center expands and exerts 258 a stronger influence on the western US, increasing the frequency of atmospheric 259 stagnation (Wang and Angell, 1999). Combining the low wind speed and low 260 probability of precipitation during stagnation with low antecedent soil moisture 261 condition generally prevalent during summer, heat waves can develop to create a 262 maximum center of combined extreme events beyond the coastal mountain ranges of the western US (Zhao and Khalil, 1993). The eastern central US is prone to heat wave 263 264 and stagnation as a result of the upper level ridge that develops during summer in that 265 region. These climatic conditions give rise to the dipole patterns of maximum heat wave 266 and stagnation in the western and eastern central US. The dipole pattern becomes more obvious and magnified for the compound events because stagnation can promote the 267

268 development of heat waves, as discussed earlier. For the compound events, the 269 simulation performs well and even better than the metrics of atmospheric stagnation 270 events. The high values in western and southeastern US, as well as the low values in 271 the central and upper Midwestern US are reasonably captured by the model, with 272 statistically significant correlation (R=0.58).

273 Thus, WRF/Chem in general well reproduced the spatial patterns and frequency 274 of the extreme weather events including heat waves, atmospheric stagnation, and their 275 compound events. Although atmospheric stagnation occurs more than 20 days during 276 the summer in large areas over the western and eastern US, heat waves do not occur for 277 more than 10 days generally, so the compound events of heat waves and stagnation are 278 rather rare and occur on average for no more than 5 days during summer over the US. 279 In the next section, ozone concentrations during these extreme weather events are 280 analyzed.

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3.2 Evaluation of ozone concentrations during extreme weather events 282 283

284 Maximum daily 8-hr (MDA8) ozone is an important variable considering its close 285 relationship with human health (USEPA, 2007) so we focus on the evaluation of MDA8 286 O₃ during summertime. Fig. S1 shows the spatial distributions of MDA8 ozone with or 287 without extreme weather event in the WRF/Chem simulations and the NARR/AQS 288 observations. MDA8 ozone with extreme weather events (Fig. S1; left panels) show 289 similar increase compared to MDA8 ozone without extreme weather events in both 290 model simulations and observations over the eastern US. In the west coast, the increase 291 is slightly higher in model simulations than in observation. Overall, WRF/Chem well 292 reproduced the influence of extreme weather event on enhancing MDA8 ozone over 293 the US.

294 From the perspective of public health, USEPA (2007) recommended attention to 295 ozone values higher than 40 ppbv because the human impact of ozone is small for low 296 ozone concentration. Thus, we compare the mean ozone concentrations during summer of 2001-2010 between observed data (AQS) and model results for the following three conditions in Fig. 3: 1) days with heat waves, but no atmospheric stagnation; 2) days with atmospheric stagnation but no heat waves; 3) days with compound events (both heat wave and atmospheric stagnation) occurring. Thus the first two conditions identify single extreme events and the third condition identifies compound extreme events. We compare observed ozone concentration greater than or equal to 40 ppbv and the simulated ozone concentration corresponding to the same locations of the observations.

304 As depicted in Fig. 3, WRF/Chem reasonably reproduced the observed ozone 305 concentrations during the extreme weather events, showing statistically significant 306 correlations with the observed AQS data. Moreover, if the benchmark (15%/35% for 307 MFB/MFE and 10%/20% for NMB/NME) suggested by USEPA (2007) is used as a 308 reference, all the statistical metrics based on evaluation against ozone higher than 40 309 ppbv in observations are within or much smaller than the benchmarks, illustrating 310 promising ability of WRF/Chem in simulating the ozone concentrations during heat 311 waves, stagnation, and their compound events. Even if all ozone values including values 312 below 40 ppbv are considered, the four metrics (MFB/MFE and NMB/NME) are mostly 313 within the benchmarks and the correlation coefficients between model and observation 314 are only slightly reduced by 0.04, 0.11, and 0.1 for the three types of extreme weather 315 events, respectively, and all values are still statistically significant. However, the 316 general low biases of the simulations are obvious from the regression lines. Ozone 317 concentrations during compound extreme events are clearly shifted to higher values 318 relative to ozone concentrations during single extreme events.

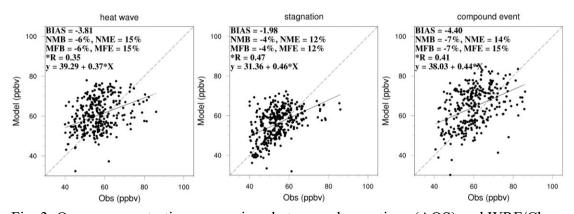


Fig. 3. Ozone concentration comparison between observations (AQS) and WRF/Chem simulations during heat waves (left), atmospheric stagnation (middle), and compound heat wave and atmospheric stagnation events (right). Metrics shown inside each figure were from formula (A1) to (A6) in the Appendix. An r-test (a=0.05) is performed to test the statistical significance and *R indicates statistical significance at 95% confidence level. The solid line is the linear regression line, and the dashed line is a one-to-one reference line.

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329 To delve into the spatial heterogeneity, ozone concentrations from model and 330 observations for the three types of extreme weather events are shown using box-and-331 whisker plots in Fig. 4. Considering the detrimental effect on human health when 332 MDA8 ozone concentration exceeds 70 ppbv by National Ambient Air Quality Standards (NAAQS), we evaluate the WRF/Chem simulated ozone concentrations 333 334 above this particular threshold. We calculated the mean values of MDA8 ozone 335 concentration exceeding 70 ppbv for each type of extreme weather events, and the mean 336 values are marked at the top of each panel in Fig. 4.

337 The box-and-whisker plots show some unique features in the observations. For 338 example, the mean ozone (red dot) concentrations tend to be slightly higher when heat 339 waves and stagnation occur at the same time, while the mean values are relatively lower 340 during atmospheric stagnation than during heat waves. These are consistent with Fig. 3 when values are plotted regardless of the regions. This feature was reasonably captured 341 342 by the model, in particular over regions in the eastern US, such as Northeast and 343 Southeast. Regarding high ozone concentrations (i.e., values higher than 70 ppbv), the 344 model is skillful in the eastern US with major anthropogenic emissions. The mean bias 345 could be as small as 0.4 ppbv (over the Southeast during heat waves), and mostly within 1 ppbv. However, for some regions, i.e., West and Southwest, negative biases could reach a few ppbv; the negative biases in many regions are likely linked to an underestimation of heat wave intensity, which is reflected in the underestimation of heat wave days as shown in section 3.1. Other possible reasons for the negative biases in surface O₃ include uncertainties in precursor emissions, boundary conditions, as well as overpredictions in precipitation, as reported in Yahya et al. (2017a).

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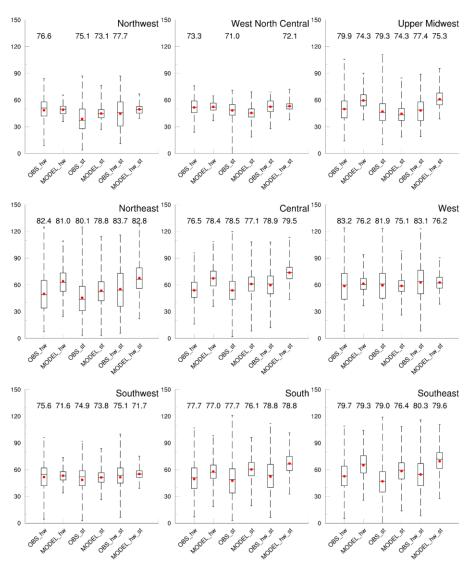




Fig. 4. MDA8 ozone concentration comparisons during the summer of 2001-2010 in nine climate regions (according to Fig. 1), with box-and-whisker plots showing the minimum, maximum (line end-points), 25th percentile, 75th percentile (boxes), medians (black lines) and average (red point) of mean MDA8 ozone from observation (NARR/AQS; with prefix OBS_) and model (WRF/Chem; with prefix MODEL_)

during heat waves (with suffix hw), atmospheric stagnation (with suffix st) and
compound events of both heat wave and atmospheric stagnation (with suffix of hw_st).
The numbers at the top of each panel indicate the average values of MDA8 ozone
concentration above the standard (70ppbv).

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To further evaluate the capability of WRF/Chem in modeling high ozone (beyond 365 366 70ppbv), Fig. S2 displays the interannual variability of high ozone over the US in the WRF/Chem simulations and AQS observations. For observations, the variance of 367 368 annual mean high ozone were calculated only for grids with more than five years of 369 data. Similar to the ozone distribution in Fig. S1, larger values are mainly found in the 370 west coast and the eastern and central US. Variance over the eastern US in observations is high while WRF/Chem is in general slightly smaller. Considering the total high ozone 371 372 episodes in historical periods, the contributions of extreme weather events to the high ozone episodes are shown in Fig. S3. Only grids having 10 days or more with high 373 374 ozone are shown to avoid grid cells with very high fractions due to the small number 375 of high ozone episodes. WRF/Chem simulated a slightly larger fraction in the west coast 376 compared to observations and well captured the high fraction in the eastern US. This feature is similar to the ozone distribution in Fig. S1. Hence overall, WRF/Chem 377 378 demonstrates a reasonable capability of modeling high ozone episodes and the 379 contribution of extreme weather events to high ozone episodes in the US.

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381 4. Impacts of extreme events and climate change on ozone

- 382 concentrations
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384 4.1 Impacts of single and compound extreme events on ozone
385 concentrations

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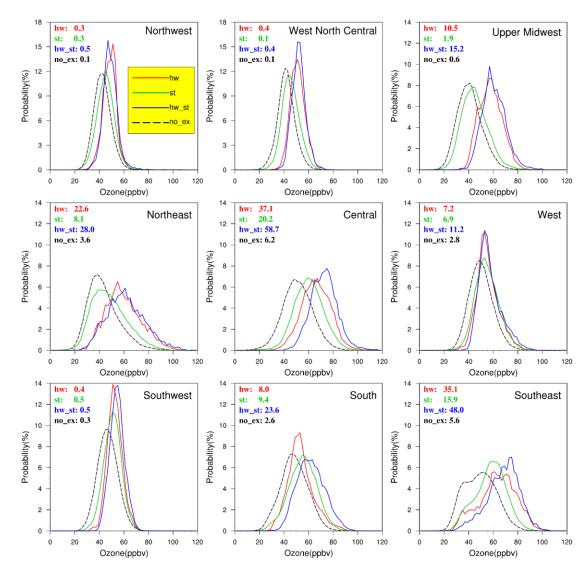
To investigate the impacts of the extreme weather events on ozone concentrations,
we composited the MDA8 ozone concentrations from WRF/Chem for the three types

of extreme weather events and to periods without any extreme event (non-extreme
event) in summer of 2001-2010 using probability density functions (PDFs) shown in
Fig. 5.

392 By comparing the solid lines (extreme event period) and dashed line (non-extreme 393 event period) in Fig. 5, all extreme weather events have positive impacts on ozone 394 particularly at the high-end tail of the distributions. The difference between ozone 395 concentrations with and without extreme events is statistically significant in all regions at the 95% confidence level. For regions with mean ozone values exceeding 70 ppbv 396 397 (numbers shown in Fig. 5), much larger differences are noticeable between the PDFs of 398 extreme and non-extreme periods, with extreme events notably shifting both the low-399 end and high-end tails towards higher values. These regions include Northeast, Central, 400 South, and West. Conversely, regions such as Northwest, West North Central and 401 Southwest show negligible differences between the PDFs. The spatial heterogeneity is 402 closely related to the spatial distribution of emissions in the US, i.e., regions with larger 403 increase of ozone concentration particularly near the high-end tail (i.e., Northeast, 404 Southeast, Central, Upper Midwest, South and West) due to extreme weather events are 405 also areas with higher anthropogenic emissions in the US (see also Fig. 3 in USEPA 406 (2007)). Thus, stronger photochemical reactions in those regions may enhance the 407 effect of extreme weather events on ozone formation.

Now comparing the effects of different types of extreme weather events on ozone 408 409 concentrations (solid lines of different colors in Fig. 5), the effect of heat waves on 410 ozone formation is generally larger than the effect of atmospheric stagnation, whereas 411 the compound effect is larger than the effect of either type of single extreme weather 412 event. This feature displays similar spatial heterogeneity as discussed above, i.e., the 413 largest impact from the compound effect occurs in the South and Central (about half of 414 the compound events leading to MDA8 ozone higher than 70 ppbv), followed by 415 Northeast, South, Upper Midwest and West (11%-28% compound event days resulting 416 in MDA8 O₃ of 70 ppbv or higher) and negligible increase from the compound events

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421 Fig. 5. Composited probability density distributions of MDA8 ozone simulated by 422 WRF/Chem for three types of extreme weather events (solid lines) and non-extreme event periods (dashed line) during summer of 2001-2010 in nine regions (according to 423 424 Fig. 1). Each panel includes four numbers on the upper left showing the probability of 425 MDA8 ozone higher than 70ppbv during extreme weather events for heat waves (hw:red), stagnation (st:green), compound extremes events (hw st:blue) and non-426 extreme periods (no ex:black). Note that all panels except for the Northwest and West 427 428 North Central use the same scale for the y-axis

Besides the distinguishing impacts extreme events have on ozone relative to nonextreme days, how high the concentration of ozone can reach during extreme events may depend on the intensity of the extreme events and the emissions. Fig. 6 shows the 433 correlations between ozone concentration with the daily maximum 2-meter temperature 434 during heat waves and 10-meter wind speed during atmospheric stagnation events. The 435 correlations between temperature and ozone are positive and statistically significant in 436 areas with high emissions such as Northeast, Central, Upper Midwest, South, and 437 Southeast. For stagnation events, the correlations are statistically significant mainly in 438 South, Southeast, and along the west coast. These correlations between ozone and the 439 intensity of extreme events are consistent with the shift of the high-end tails of the PDFs 440 to higher ozone values, as shown in Fig. 5. In areas with low emissions (e.g., Northwest 441 and West North Central), ozone concentrations are not well correlated with the intensity 442 of extreme events because the production of ozone is limited by the low emissions 443 (Vingarzan, 2004). Hence only the low-end instead of the high-end tails of the PDFs 444 are shifted to higher values in regions with low emissions, and the PDFs on extreme 445 days are noticeably narrower compared to the PDFs on non-extreme days (Fig. 5). As 446 climate change may increase the frequency as well as the intensity of extreme events, 447 ozone concentrations may be affected, regardless of emissions control in the future. 448

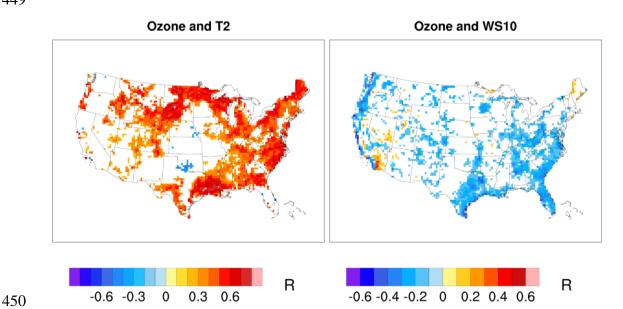


Fig. 6. Correlation between ozone concentration and (left) daily maximum 2-meter
temperature (T2) during heat waves and (right) 10-meter wind speed (WS10) during
atmospheric stagnation in the WRF/Chem simulations. Only values that pass the t-test

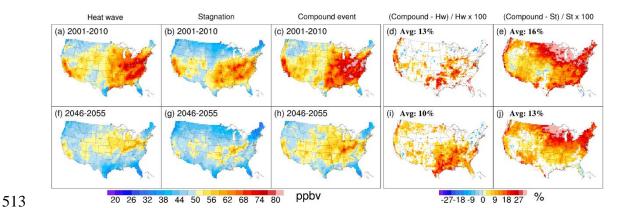
456 **4.2 Impacts of climate change on ozone concentrations**

457

Having investigated the impacts of extreme weather events on ozone 458 459 concentration, we now focus on how ozone concentrations may change in the future 460 with climate change, changes in biogenic emissions in response to changes in climate, 461 and large anthropogenic emission reductions in the RCP 8.5 scenario. Fig. 7 shows the 462 spatial variations of ozone concentrations composited during extreme weather events 463 at present (top row) and in the future (bottom row). The spatial features displayed in 464 the top row are in agreement with what have been observed from Fig. 5, showing larger 465 impacts of extreme weather events on ozone formation east of the Rockies for both 466 single extreme events and compound events (Figs. 7a,b,c). Similarly large impacts are 467 also found in California, which are obscured in the regional average shown in Fig. 5. 468 Averaged over the US, MDA8 ozone concentrations increase by 22% and 12% during 469 heat waves and stagnation events compared to non-heat wave and non-stagnation days. 470 Compound events have significantly higher impact on ozone compared to the single 471 extreme events, with statistically significant differences of 13% and 16%, respectively, 472 for heat waves and stagnation (Figs. 7d,e). To understand why compound events have 473 larger impacts than single extreme events, Fig. S4 shows that during compound event 474 days, the daily maximum 2-meter temperature is comparable to that during heat waves 475 but 6.27°C higher than that during stagnation events, leading to a 16% increase in 476 MDA8 O₃ during compound events relative to stagnation events. Similarly, the 10-477 meter wind speed during compound events is comparable to that during stagnation events but 1.4 ms⁻¹ weaker than during heat wave days, leading to a 13% increase in 478 479 MDA8 O₃ relative to heat wave days.

In the future, as anthropogenic emissions are projected to decrease substantially (i.e., Table 2 in USEPA (2007)), the mean ozone concentration correspondingly decreases during both single extreme events and compound events compared to the present day (i.e., Figs. 7f,g,h vs. Figs. 7a,b,c). However, even with the dramatic 484 anthropogenic emission reduction (i.e., 50% or more reduction in non-methane volatile 485 organic compounds and nitrogen oxides based on Table 2 in Gao et al. (2013)), extreme 486 weather events can still trigger the formation of high ozone concentration (e.g., in 487 central eastern US in Figs. 7f,g,h) to reach or exceed the present-day national standard 488 of 70 ppbv. From Fig. S4, the daily maximum 2-meter temperature is 5.54°C warmer during compound events than stagnation events, leading to a 13% increase in MDA8 489 490 O₃ during compound events relative to stagnation events. Similarly, the 10-meter wind speed is 1.28 ms⁻¹ weaker during compound events than heat wave events so MDA8 O₃ 491 492 increases by 10% during compound events relative to heat wave events in the future. 493 Hence, compound events increase ozone concentrations by 10% and 13% more than 494 the effect of heat wave only and stagnation only, respectively. These numbers shown in 495 Figs. 7i, j are only 3% lower than those of the present day (Figs. 7d,e).

496 Despite dramatic reduction in anthropogenic emissions in the RCP 8.5 scenario 497 (Riahi et al., 2011), extreme weather events are still important considerations for air 498 quality and health in the future. This is because both frequency and intensity of extreme 499 events increase in the future, which compensate partly for the effects of reduced 500 emissions. From Fig. S5, heat waves occur on average 13.67 days more and 0.98°C 501 warmer in the future relative to the present, with most of the increase occurring in the 502 western US. There is no increase in the number of stagnation days in the future when 503 averaged over the US (Fig. S5), and the change in wind speed during stagnation is also 504 negligible (Fig. S6). However, the daily maximum 2-meter temperature is 1.42°C 505 warmer during stagnation events in the future compared to the present (Fig. S5). Lastly, 506 compound events occur on average 4.91 days more often, with temperature 1.25°C 507 warmer in the future compared to the present (Fig. S5). Hence the increase in the 508 number of heat waves and the warmer temperature during heat waves as well as 509 stagnation events increase their individual and compound effects on ozone 510 concentrations in the future. These motivate analysis of changes in extreme events in 511 the future using a multi-model ensemble for more robust results.



514 Fig. 7. Spatial distributions of mean MDA8 ozone concentrations simulated by 515 WRF/Chem for three types of extreme weather event episodes and the relative 516 difference between compound event and single event during summer in 2001-2010 (top 517 row) and 2046-2055 under RCP 8.5 (bottom row). In (d,e,i,j), only values with 518 statistically significant differences (t-test: a=0.05) between the compound effect and 519 single event are shown, and the mean differences are labelled on the top left.

- 520
- 521

5. Changes of extreme weather events in future by CMIP5

522

523 To provide further insight of future changes in ozone concentration, we analyzed 524 changes in extreme weather events using the multi-model ensemble of CMIP5 data. 525 Using CMIP5 data complements our analysis of the WRF/Chem simulations in two ways. First, CMIP5 model outputs are available for a continuous period through 2100. 526 527 We analyzed three time periods, each 20 years long, for 1991-2010 as historical period, 528 and 2041-2060 and 2081-2100 in RCP 8.5 as future periods. Extending the analysis 529 period from 10 years for the regional climate simulations to 20 years for CMIP5 allows 530 for a more statistically robust analysis of extreme events. The added period of the late 531 century, 2081-2100, will elucidate how extreme weather events evolve with continuous 532 warming. Second, we extended our analysis using CMIP5 data to the entire northern 533 hemisphere starting from 20°N. The inclusion of other continents such as Europe and China provides useful information for how extreme weather events may change in 534 densely populated regions, with potential impacts on air quality and health. Analysis of 535 536 the CMIP5 mean extreme event days over the US shows that in general, the CMIP5

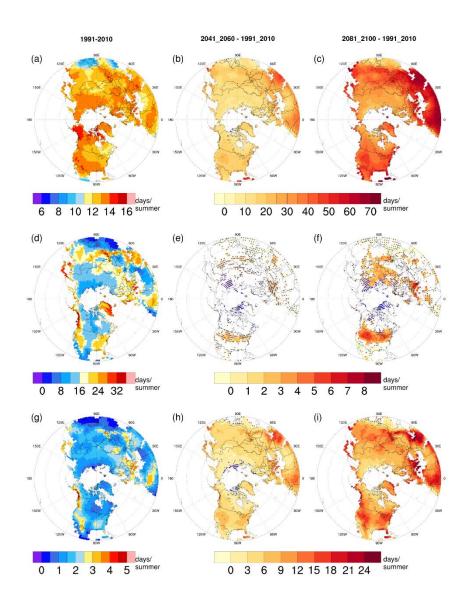
mean has spatial patterns comparable to those of the observations and WRF/Chem simulations but it has a much lower number of extreme event days, especially for stagnation and compound events (not shown). The CMIP5 mean projected changes in extreme event days also show comparable spatial patterns those of WRF/Chem over the US, but again, the magnitudes of change are much smaller (not shown). Analysis of the CMIP5 projections of extreme event changes is important to provide a multi-model context of uncertainty.

544 The summer mean number of days at present (1991-2010) and changes in future 545 (2041-2060, 2081-2010) for heat waves, atmospheric stagnation, and compound events 546 are shown in Fig. 8. For robust comparisons between future and present climate, both 547 model agreement and significance are considered, as adopted by previous studies (Gao 548 et al., 2013; Gao et al., 2014). A total of 20 models were selected (listed in Table 1), and 549 values at any grid cell are considered to have agreement if more than 70% of the models 550 agree with the CMIP5 mean on the sign of the change. Once agreement is established, 551 statistical significance is tested over the grid cells, and the values at any grid cell are 552 statistically significant if at least half of the CMIP5 models show statistical significant 553 changes (t-test, a=0.05). After the tests, most of the grid cells showing model agreement 554 also passed the statistical significance test; blue dots indicate grid cells with no 555 significant changes of extreme weather events. Three major continents were selected 556 for analysis and the results are summarized in Table 2.

557 As shown in Fig. 8 and Table 2, at present (Figs. 8a,d,g), the mean annual numbers 558 of heat waves, atmospheric stagnation and compound events are 12.9, 16.4 and 1.6, 559 respectively. In the future, there are robust increases of heat wave days worldwide, 560 consistent with previous studies (Seager et al., 2013), with a mean increase around 200% 561 by the end of this century. The changes in atmospheric stagnation are in general smaller 562 than the changes in heat waves; however, large increases can also be found in some 563 areas such as the western US. This is in contrast with the insignificant change in 564 stagnation days from the WRF/Chem simulation (Fig. S5), demonstrating the

565 importance for using a multi-model ensemble and investigating changes not just in the mid-century but further towards the end of the century when climate change signals 566 567 become more prominent (Figs. 8e,f). The overall increase in stagnation events is on 568 average 1 day per summer in the future over the northern hemisphere for atmospheric stagnation by the end of this century. Moreover, it is obvious that the compound event 569 570 shows more dominant increases than stagnation event, with 2 days or less at present on 571 average, but more than 10 days on average in the US, Europe and China. Since we have 572 demonstrated that compound events have larger impact on ozone than single extreme 573 events (Fig. 5), the large increase in compound event days suggests that they will be 574 important considerations for projecting high ozone episodes.

575



577 Fig. 8. Spatial distribution of historical (left column) and future changes in the mid-578 century (second column) and end-of-century (third column) in the number of extreme 579 weather days per summer for heat waves (top row), atmospheric stagnation (middle 580 row) and compound events (bottom row) from CMIP5 over land in the north 581 hemisphere north of 20° N. For the future changes, only grids showing model 582 agreement are shown, with blue dots representing values with no statistical 583 significance.

584

585 As discussed in Section 4, both the frequency and intensity of extreme events have

586 important effects on ozone concentrations. From Fig. S7, the intensity of heat waves

587 is projected to increase with time throughout the 21^{st} century as warming increases.

588 Both the WRF/Chem and CMIP5 results show larger increase in heat wave intensity

589 in the western US. During stagnation and compound events, the daily maximum 2-

590 meter temperature also increases with time. Consistent with WRF/Chem results (Fig.

591 S6), CMIP5 also shows negligible changes in wind speed during atmospheric

592 stagnation and compound event, but decrease during heat waves (Fig. S8), further

- 593 enhancing the effect on ozone formation.
- 594

Table 2. Average number of days of extreme weather event episodes in summer of 1991-2010, 2041-2060 and 2081-2100, along with the future increase over the northern hemisphere (NH) and three regions including the United States (US), Europe, and China. Statistical significance test was applied using a t-test (α =0.05), and values with no statistical significance are italicized.

	Heat wave (days/summer)			
Areas	Hist (1991~2010)	2041~2060 - Hist	2081~2100 - Hist	
NH	12.9	15.6	36.5	
US	13.3	17.3	39.7	
Europe	13.1	16.0	37.8	
China	12.3	16.3	39.2	
A #200	Stagnation (days/summer)			
Areas	Hist(1991~2010)	2041~2060 - his	2081~2100 - Hist	
NH	16.4	0.2	0.9	
US	18.0	0.6	1.7	
Europe	21.9	0.2	0.9	
China	17.4	0.1	0.6	

	Compound events (days/summer)			
Areas	Hist (1991~2010)	2041~2060 - Hist	2081~2100 - Hist	
NH	1.6	4.1	9.2	
US	2.0	5.1	11.3	
Europe	1.9	4.9	11.5	
China	1.6	4.6	10.5	

602 6. Conclusions and Discussions

603

604 The regional model WRF/Chem version 3.6.1 has been used to downscale simulations from the CESM NCSU global model. The regional model well reproduced 605 606 the frequency of extreme weather events, including heat waves, atmospheric stagnation 607 and their compound events, and the ozone concentration during these extreme weather 608 events at present, compared to observations. Through comparison of ozone 609 concentrations during extreme weather events period and non-extreme period, we 610 established statistically significant higher ozone concentrations during the extreme 611 event period. In particular, compound events yield the highest contribution to high 612 ozone formation, followed in general by heat waves and atmospheric stagnation.

Compound events have larger impacts on ozone than single events because the 613 614 temperature during compound events is noticeably higher than that during stagnation-615 only events and the wind speed during compound events is noticeably weaker than 616 during heat wave-only events. The combination of warmer temperature and weaker 617 winds promote photochemical reactions that produce high ozone episodes. Also importantly, ozone concentrations increase with the intensity of extreme events in 618 619 regions with high emissions, leading to a shift in the PDFs towards higher ozone values, 620 and increasing the frequency of occurrence of high ozone episodes. In regions with low 621 emissions, extreme events noticeably increase the ozone concentrations at the low-end 622 tails, but the high-end tails are not shifted, leading to narrower PDFs during extreme 623 events relative to non-extreme events.

624

In the future, under the RCP 8.5 scenario, albeit large reductions in anthropogenic

625 emissions projected, extreme weather events can still trigger the formation of higher 626 ozone concentration. The increase in ozone concentrations during extreme events 627 relative to non-extreme events is comparable in the future as in the present. Furthermore, 628 compound events of heat waves and stagnation continue to have larger impacts on 629 ozone concentrations relative to the single weather extreme events. By utilizing a total 630 of 20 CMIP5 models, we found that under climate warming, more frequent extreme 631 weather events are projected to occur in mid- to end of this century. Among the 632 increases by the end of the century, compound events show a dominantly higher 633 fractional increase by a factor of 4-5, compared to the single events, i.e., heat waves (~ 634 a factor of 2) or atmospheric stagnation ($\sim 14\%$), as shown in Table 2.

635 Since the CMIP5 models do not include detailed atmospheric chemistry, we cannot assess how ozone concentrations may change in the mid-to-late 21st century. The 636 CMIP5 results indicate robust increases in the frequency and intensity of heat waves 637 638 and frequency of compound events with higher temperature in the future. While 639 reductions of anthropogenic emissions in the RCP 8.5 scenario will likely counter the 640 effects of extreme events on ozone concentrations, the frequency of high ozone 641 concentrations is enhanced by extreme events even in low emission regions (e.g., 642 Northwest) in the present day (Fig. 5). Hence it is likely that high ozone episodes may 643 still occur in the future due to increases in extreme heat, despite reductions in 644 anthropogenic emissions, with adverse effect to human health.

645 However, similar to how low emissions constrain the high-end tails of the PDFs 646 of ozone from shifting to very high or extreme ozone concentrations even under 647 extreme weather conditions (e.g., Northwest in Fig. 5), reductions in anthropogenic 648 emissions in the future could reduce or eliminate the occurrence of extreme high ozone 649 episodes. Hence controlling anthropogenic emissions may be critical for reducing the 650 impacts of extreme events on extreme air quality episodes and associated human health 651 impacts. This may be especially important in regions like China that have experienced severe air pollution in the recent decades. More attention to improving projections of 652

653 compound events and evaluating their impacts on ozone may better constrain the projections of extreme air quality episodes and inform strategies to reduce their 654 655 detrimental effects on human health now and in the future.

656

Appendix 657

658

659 Statistically metrics for evaluating model performance

660

Metrics for model performance evaluation used in this study include BIAS (Mean 661 Bias), NMB (Normalized Mean Bias, percent), NME (Normal Mean Error, percent), 662 663 MFB (Mean Fractional Bias, percent), MFE (Mean Fractional Error percent) and R (Correlation Coefficient). Calculations of these metrics are shown below in Eqs. (A1)-664 (A5), where N is the number of sample size, MODEL and OBS represent the 665 corresponding value in model simulation and observation (AQS sites or reanalysis data), 666 667 respectively. As low OBS values can amplify the metrics, a cutoff of 40 ppbv or 60 ppbv of ozone is suggested in evaluation for ozone. Benchmarks of MFB and MFE for 668 669 O₃ are 15% and 35%, and of NMB and NME for O₃ are 10% and 20% (Tebaldi et al., 2011). 670

671
$$BIAS = \frac{1}{N} \sum_{1}^{N} (Model - Obs)$$
(A1)

672
$$NMB = \frac{\sum_{i=1}^{N} (Model - Obs)}{\sum_{i=1}^{N} (Obs)} \times 100\%$$
(A2)

673
$$NME = \frac{\sum_{i=1}^{N} |Model - Obs|}{\sum_{i=1}^{N} (Obs)} \times 100\%$$
(A3)

674
$$MFB = \frac{2}{N} \sum_{1}^{N} \left(\frac{(Model - Obs)}{(Model + Obs)} \right) \times 100\%$$
(A4)

675
$$MFE = \frac{2}{N} \sum_{1}^{N} \left(\frac{|Model - Obs|}{(Model + Obs)} \right) \times 100\%$$
(A5)

676
$$R = \frac{\sum_{1}^{N} (Model - \overline{Model})(Obs - \overline{Obs})}{\sqrt{\sum_{1}^{N} (Model - \overline{Model})^{2} \sum_{1}^{N} (Obs - \overline{Obs})^{2}}}$$
(A6)

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