

1 **TITLE:**

2 **Exacerbated ozone pollution in the greening northern China**

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32 **Exacerbated ozone pollution in the greening**  
33 **northern China**

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47 Key words: ozone, afforestation projects, climate warming, air pollution in  
48 cities

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52 **SUMMARY**

53 Biogenic volatile organic compounds are vital precursors of tropospheric ozone  
54 ( $O_3$ ). In the past decades, northern China has witnessed the strongest  
55 enhancement of vegetation covers due to national afforestation projects;  
56 however, response of the vegetation greenness to near-surface  $O_3$  remains  
57 unclear. By integrating measurements and numerical simulations, here we  
58 show that  $O_3$  pollution in northern China has been enhanced by the unwind  
59 greening over the past two decades and will be exacerbated further in this  
60 century as the climate warming. We found that isoprene emission increased by  
61 30% over northwestern China because of the vegetation increase and regional  
62 warming. Together with changes in anthropogenic emissions, it caused more  
63  $O_3$  pollution in city clusters in northern China. In the context of carbon peaking  
64 and carbon neutrality policy, vegetation greening will cause aggravating  $O_3$   
65 pollution by the middle of this century, which need to be considered in future air  
66 quality measures in China.

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## 72 INTRODUCTION

73 Vegetation greenness has been increasing globally in past decades, partly  
74 due to the global warming and carbon dioxide (CO<sub>2</sub>) fertilization<sup>1-4</sup>. At regional  
75 scale, extensive reforestation and afforestation has been conducted to address  
76 the challenge of climate change in terms of carbon capture and eroded<sup>5-7</sup>.  
77 Therefore, the greening rate shows strong regional diversity, with the most  
78 pronounced greening over intensively farmed or afforested areas, particularly  
79 in the Eurasia continent<sup>8</sup>.

80 The northwestern China has witnessed the most intensive afforestation  
81 activities and featured the largest, around 2 million ha per year, afforested area  
82 in the world<sup>9</sup>. Such a fast vegetation greening is predominantly attributed to a  
83 series of large-scale afforestation program implemented by the Chinese  
84 government for the purpose of alleviating land degradation<sup>10</sup>. Noteworthy, the  
85 Three Northern Shelter Forest (TNSF) project, known as “the Green Great Wall”,  
86 increased the vegetation cover of the Gobi Desert from 5.05% to 13.2% from  
87 1977 to 2016. Similarly, the Grain for Green project doubled the vegetation of  
88 the Loess Plateau since 1999<sup>11-13</sup>. Such a change of land use and land cover  
89 has been found a strong impact to regional climate in these region<sup>13-16</sup>.

90 Vegetation greenness not only feedbacks to the local and regional climate,  
91 but also would greatly reshape the atmospheric chemistry via biological  
92 processes<sup>1,17,18</sup>. The biogenic volatile organic compounds (BVOCs), including

93 isoprene and terpenes, from terrestrial vegetation play an important role in  
94 atmospheric oxidative balance and the lifetimes of trace gases<sup>19,20</sup>. Due to the  
95 strong chemical reactivity, BVOCs, once released, would react with ambient  
96 oxidants such as the hydroxyl radical (OH) and subsequently contribute to  
97 ozone (O<sub>3</sub>) and other oxidization products<sup>21</sup>. Vegetation emission has been  
98 found to greatly deteriorate O<sub>3</sub> pollution by supplying sufficient precursors for  
99 photochemical reactions, particularly when it mixed with anthropogenic  
100 pollutants like nitrogen oxides (NO<sub>x</sub>)<sup>22-25</sup>.

101 Ozone pollution is currently one of the key environmental challenges in  
102 China despite widely-implemented emission control measures<sup>26,27</sup>. In northern  
103 China, a hotspot with intensive anthropogenic emission, what role vegetation  
104 greenness has played in O<sub>3</sub> pollution during past decades remains unclear<sup>28</sup>.  
105 Most notably, Chinese government has launched a long-term plan of to  
106 increase forest cover to 26% by 2050<sup>29</sup>. How the future aggressive afforestation  
107 in a warming climate will influence regional O<sub>3</sub> pollution is unknown. In this study,  
108 by integrating satellite retrievals, in-situ observations, and climate-chemistry  
109 coupled modelling as well as future climate projections, we provide a  
110 quantitative understanding on the role of vegetation greening in atmospheric  
111 chemistry and O<sub>3</sub> pollution in the fast-greening northern China in the past  
112 decades and future.

## 113 RESULTS

### 114 Vegetation greening and ozone increase in northern 115 China

116 Since the 1980s, multiple ecological afforestation projects have been  
117 carried out to prevent environmental deterioration in the semi-arid regions of  
118 northern China<sup>8,30</sup>. Affected by these projects, notable growth of vegetation  
119 greening occurred in the Loess plateau, where the summertime leaf area index  
120 (LAI) and the Normalized Difference Vegetation Index (NDVI)<sup>31,32</sup> increased by  
121 almost 77% and 46%, respectively, from 2001 to 2019 (Figure. 1a; Figure. S1).  
122 Under such a fast greenness expansion, it is expected that emission of BVOCs  
123 and O<sub>3</sub> concentration would consequently increase in the Loess plateau,  
124 especially in warm seasons (Figure. S2a). Indeed, ground-based observations  
125 from air quality monitoring network in China<sup>33</sup> show that near-surface O<sub>3</sub>  
126 concentration in northern China rises throughout the year with highest trend in  
127 summer (Figure. S2b). Over the fast-greening Loess Plateau and its downwind  
128 areas, near-surface O<sub>3</sub> concentrations increased by 8–24 ppb from 2013 to  
129 2019 (Figure. 1b). The regional-scale O<sub>3</sub> enhancement cannot be explained  
130 alone by the change in anthropogenic emission<sup>34</sup>, as which tends to cause  
131 different responses in urban and remote areas<sup>35</sup>. Emission estimation suggests  
132 a 23.6% reduction in NO<sub>x</sub> emission together with a tiny change in anthropogenic  
133 volatile organic compounds (VOCs) emission, which cannot explain the strong

134 O<sub>3</sub> change in the Loess Plateau, a NO<sub>x</sub>-limited area<sup>36</sup> (Figures. S3 and S4a). In  
135 addition, tropospheric ozone derived from the Ozone Monitoring Instrument  
136 (OMI)<sup>37</sup> increase continuous by 0.3 DU a<sup>-1</sup> in northern China from 2005 to 2019,  
137 indicating drivers other than reduction of anthropogenic emissions, as the later  
138 only decreased since 2013.

139

## 140 **Enhanced interaction of anthropogenic and biogenic** 141 **sources**

142 To explore the underlying mechanism of the soaring near-surface ozone  
143 over a greening northern China, we conducted meteorology-chemistry coupled  
144 model simulations for the past two decades. Model predictions were evaluated  
145 using ambient data at 317 air quality monitoring stations. As shown in Figure.  
146 S4b, the model well reproduced the observed spatial and temporal variations  
147 of O<sub>3</sub> concentrations in northern China. To investigate whether the afforestation  
148 project contributes to the rising O<sub>3</sub> pollution, we examined the impact of  
149 increased BVOC emissions (mainly isoprene) on the maximum 8-hour-average  
150 90-percentile (M8A90) O<sub>3</sub> concentrations in summer (Figure. 2a). The M8A90  
151 O<sub>3</sub> continued to exceed the Chinese National Ambient Air Quality Standard of  
152 82 ppb<sup>26</sup> in northern China, which is recommended by the Ministry of Ecology  
153 and Environment China to characterize the statistic potential severe damage to  
154 human health<sup>33</sup>.

155 Changes in emissions and climate are the two main factors that drive the  
156 variations in surface O<sub>3</sub> concentrations<sup>38</sup>. For the entire period 2001–2019, the  
157 emission of BVOCs around the Loess Plateau is expected to increase by 30%  
158 related to the afforestation alone. However, simulations using fixed  
159 meteorology and anthropogenic emissions of 2010 together with increasing  
160 BVOCs (GREEN) resulted in only a 2% increase in local M8A90 O<sub>3</sub>  
161 concentrations in the fastest greening area (Figure. 2a). The relatively low trend  
162 could be explained by the NO<sub>x</sub>-limited regime over the Loess Plateau (Figure.  
163 S4a). However, the M8A90 O<sub>3</sub> concentrations increase more rapidly in city  
164 clusters in the North China Plain, east to the Loess Plateau (Figure. 2a).

165 It is known that isoprene can be oxidized within a few hours, while the  
166 oxidation products of isoprene, e.g. ISPD (lumped methacrolein, methyl vinyl  
167 ketone, methyl glyoxal, etc) and formaldehyde (HCHO)<sup>39</sup>, have a much longer  
168 lifetime than isoprene itself, which can be transported thousands of kilometers  
169 away downwind from the biogenic source area (Figure. 2b). The satellite-  
170 measured HCHO to nitrogen dioxide (NO<sub>2</sub>) ratios show a shift from a NO<sub>x</sub>-  
171 limited regime for O<sub>3</sub> production in rural areas in the west to a VOCs-limited  
172 regime in the city clusters in the east (Figure. 2c). Therefore, those transported  
173 biogenic oxidized products, including HCHO, methyl vinyl ketone, methacrolein,  
174 methyl glyoxal, enhancing O<sub>3</sub> production under VOCs-limited conditions,  
175 resulting in a maximum increase of 7 ppb and more than 5 O<sub>3</sub> pollution events  
176 in urban areas (Figure. 2b-c).



177 Because of the vital roles of meteorological parameters on ozone  
178 production, climate change also affects surface O<sub>3</sub> concentrations via its impact  
179 on precursor gases, chemical environment associated with O<sub>3</sub> production and  
180 loss, and transport fluxes<sup>40-46</sup>. China is warming by more than 0.5 °C per  
181 decade<sup>47,48</sup>, which could enhance biogenic emissions<sup>49</sup>. To investigate the  
182 effects of climate-warming-related biogenic emissions other than circulation  
183 changes, we conducted a simulation with the fixed meteorological fields in the  
184 year 2010 and varied BVOC emissions following the real climate change  
185 between two different periods, 2001–2003 and 2017–2019  
186 (GREEN\_CLI\_EMIS). Isoprene emissions around the northern China are  
187 projected to increase by 174 Gg during the two decades. While the significant  
188 vegetation greenness between 2001 and 2019 contributed 80% to the total  
189 increment of BVOC emissions, climate change contributed the remaining 20%.  
190 The oxidation products are then increased by around 40% in the Loess Plateau  
191 (Figure. 3a-b). Our results reveal that the rapid climate warming amplifies  
192 isoprene emissions, and therefore tripling the increased M8A90 O<sub>3</sub>  
193 concentrations in downwind city clusters from 2001 to 2019 (Figure. 3c).

194 We also estimate the overall impact of climate change on O<sub>3</sub>  
195 concentrations between the two periods (GREEN\_CLI), including combined  
196 changes in biogenic emissions, atmospheric chemistry, and transport.  
197 Compared to changes in biogenic emissions, isoprene oxidation products are  
198 reduced by 39% due to the overall effects of climate change from 2001 to 2019.

199 Meanwhile, HCHO concentrations have increased in the Loess Plateau (Figure.  
200 3b). These variations are consistent with satellite-based HCHO measurement  
201 (Figure. S4c)<sup>50</sup>, suggesting the accelerated atmospheric oxidation by climate  
202 change and a fast oxidization of isoprene into HCHO in the vegetation greening  
203 areas. With transport over hundreds of kilometers, this increased HCHO is  
204 continuously consumed and produces O<sub>3</sub> on its way to downwind city clusters  
205 (Figure. 3b). In addition, once this increased HCHO are transported to city  
206 clusters, the enhanced atmospheric oxidizing capacity also amplifies the  
207 BVOC-related O<sub>3</sub> production.

208 The M8A90 O<sub>3</sub> concentrations are estimated to increase by 5 ppb in the  
209 Loess Plateau during 2001–2019 (Figure. 3c). However, only one third of the  
210 increase is associated with changes in biogenic emissions, including the  
211 combined effects of vegetation greenness and climate warming. Instead,  
212 climate-driven change in transport is a significant driver of O<sub>3</sub> variations in those  
213 vegetation greening areas, where O<sub>3</sub> chemistry is not sensitive to biogenic  
214 emissions. In contrast, the M8A90 O<sub>3</sub> concentrations are estimated to increase  
215 by 9.6 ppb in the downwind city clusters (Figure. 3c), more than 60% of which  
216 were contributed from the vegetation greening and climate-warming-related  
217 changes in biogenic emissions. Our results highlight that climate change over  
218 the past two decades has enhanced the impact of western afforestation on O<sub>3</sub>  
219 concentrations in eastern cities, with an increase of 10% pollution events in  
220 recent years (Figure. 3d).

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## 222 **Ozone response to greening in a warming climate**

223 To understand the long-term impact of the greening in this region, we  
224 further use climate projections to conduct model simulations. The  
225 Representative Concentration Pathway (RCP) and the Shared Socioeconomic  
226 Pathway-Representative Concentration Pathway (SSP) offer a broad range of  
227 future climate change and air pollution developments, considering challenges  
228 to various levels of mitigation and adaptation<sup>51-53</sup>. In this study, RCP8.5 is chosen  
229 as the reference scenario for climate change because it assumes a fragmented  
230 world that restricts international trade in energy and technology and describes  
231 an energy-intensive, fossil-based economy<sup>54</sup>. These assumptions are  
232 consistent with current global realities and fossil fuel production plans of major  
233 producing countries<sup>55</sup>. Furthermore, RCP8.5 represents the most severe future  
234 global warming scenario and is very close to SSP5-8.5 in CMIP6<sup>56</sup>, which  
235 contains a significant climate change effect on vegetation greening.

236 As show in Figure 4a-b, when the global mean surface air temperature  
237 increases by 1.1 K from 2020 to 2050, the warming trend is more sensitive to  
238 the high latitudes in eastern China<sup>56</sup>. Therefore, the BVOC emissions from the  
239 Loess Plateau and northern China are expected to increase significantly over  
240 the next 30 years due to the afforestation projects and the ecosystem  
241 responses to climate change, respectively (Figure. 4a). In addition, the westerly

242 anomaly in 2050 will lead to more substantial transport of BVOCs and their  
243 oxidants from vegetation greening areas to downwind city clusters compared  
244 to 2020 (Figure. 4b).

245 Quantitative analyses from parallel simulations with different scenarios  
246 show that future changes in anthropogenic emissions will reduce the M8A90 O<sub>3</sub>  
247 concentrations in the Loess Plateau and city clusters by ~2-5% compared to  
248 2020 (Figure. 4c; Figure. S5). In contrast, the effect of afforestation projects  
249 alone resulting a 3.3% increase M8A90 O<sub>3</sub> concentrations in eastern cities.  
250 Therefore, while stricter air quality control policies are expected to reduce O<sub>3</sub>  
251 pollution incidents in eastern urban agglomerations such as the North China  
252 Plain, these efforts are almost offset by the vegetation greening designed to  
253 protect from a warming climate.

254 Our results suggest that future climate warming will significantly amplify the  
255 impact of afforestation project on O<sub>3</sub> pollution in city clusters in the North China  
256 Plains. The overall effects of climate change and afforestation increase M8A90  
257 O<sub>3</sub> concentrations by 8–22 ppb in the city clusters, much larger than the 2–6  
258 ppb increase in the Loess Plateau from 2020 to 2050 (Figure. S6). Considering  
259 the unfavorable chemical environment for O<sub>3</sub> generation in 2050 compared to  
260 2020 in eastern city clusters (e.g., an increase in relative humidity<sup>57</sup> and a  
261 decrease in solar radiation in eastern city clusters, Figure S7, Figure. 4c), a 3-  
262 fold increase in isoprene oxidation products such as HCHO in VOC-limited  
263 urban areas is the main reason for the increase in O<sub>3</sub> pollution (Figure. S8).

264 Compared to a previous study<sup>58</sup>, our results magnify the O<sub>3</sub> pollution from  
265 climate-driven biogenic emissions by a factor of 2, indicating an essential  
266 contribution of vegetation greening to O<sub>3</sub> pollution in urban areas under a future  
267 warming climate. Previous study reported the O<sub>3</sub> pollution events increase by  
268 ~6% in eastern China from 2017 to 2050 due to changes in future  
269 anthropogenic emissions under SSP5-8.5 scenario<sup>59</sup>. In contrast, our results  
270 show that future changes in anthropogenic emissions will slightly increase the  
271 M8A90 O<sub>3</sub> concentrations in both the Loess Plateau and city clusters in 2050  
272 under SSP5-8.5 scenario. Therefore, the afforestation projects under a  
273 warming climate may partly offset the air pollution control target, making it  
274 challenging to decrease O<sub>3</sub> pollution by reducing anthropogenic emissions.

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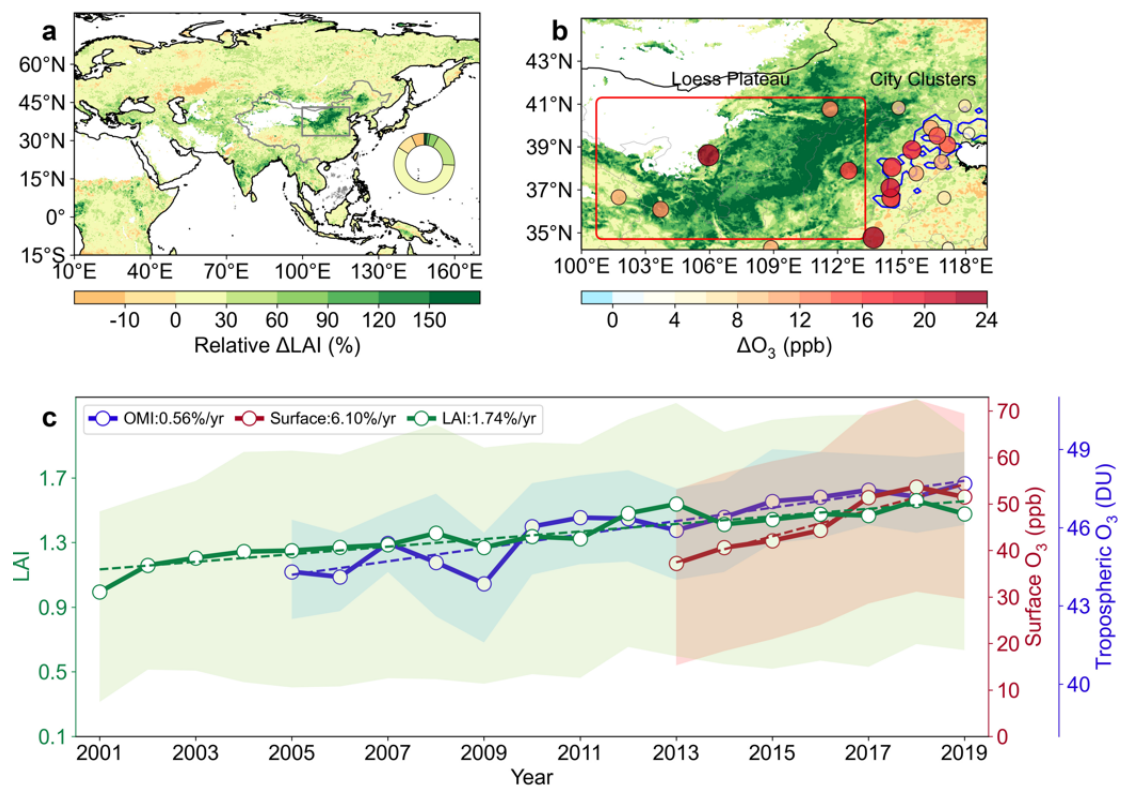
## 276 **DISCUSSION**

277 In recent decades, with the most intensive afforestation activities and the  
278 largest afforested area in the world, O<sub>3</sub> pollution in northern China has also  
279 risen sharply. In this study, we integrate a regional chemistry model WRF-  
280 CMAQ and a biogenic emissions model (MEGAN) to access how the past and  
281 future O<sub>3</sub> concentrations might change with respect to the afforestation projects  
282 and climate change. From 2001 to 2019, the combined effects of vegetation  
283 greening and climate change increased the surface regional mean M8A90 O<sub>3</sub>  
284 concentrations over the Loess Plateau and eastern city clusters by 5 and 10

285 ppb over the Loess Plateau and eastern city clusters, respectively. Although  
286 changes in O<sub>3</sub> transport fluxes were the primary reason for O<sub>3</sub> variations in  
287 vegetation greening areas, more than 60% of M8A90 O<sub>3</sub> increments in eastern  
288 city clusters were associated with changes in biogenic emissions. Compared  
289 with 2001, isoprene emissions increased by 30% in 2019 around the vegetation  
290 greening areas, and its oxidation products. Under the future global warming  
291 scenario, the contribution of the western afforestation project to O<sub>3</sub> pollution  
292 events in the eastern city clusters is even larger. The overall effects of climate  
293 change and afforestation increase M8A90 O<sub>3</sub> concentrations by 8-22 ppb in  
294 eastern city clusters.

295 To achieve carbon neutrality goals, afforestation is expected worldwide.  
296 Although anthropogenic emissions are expected to decrease in the future,  
297 aiming to reduce O<sub>3</sub> exposure in cities, afforestation projects will partially offset  
298 the benefits of air pollution control. Moreover, our results imply that the  
299 afforestation project in remote areas under a warming climate may conflict with  
300 cities' air pollution control targets. In the future, stricter air quality control policies  
301 and afforestation projects of low isoprene emission tree species<sup>60</sup> are required  
302 to meet the both challenge of climate warming and O<sub>3</sub> pollution.

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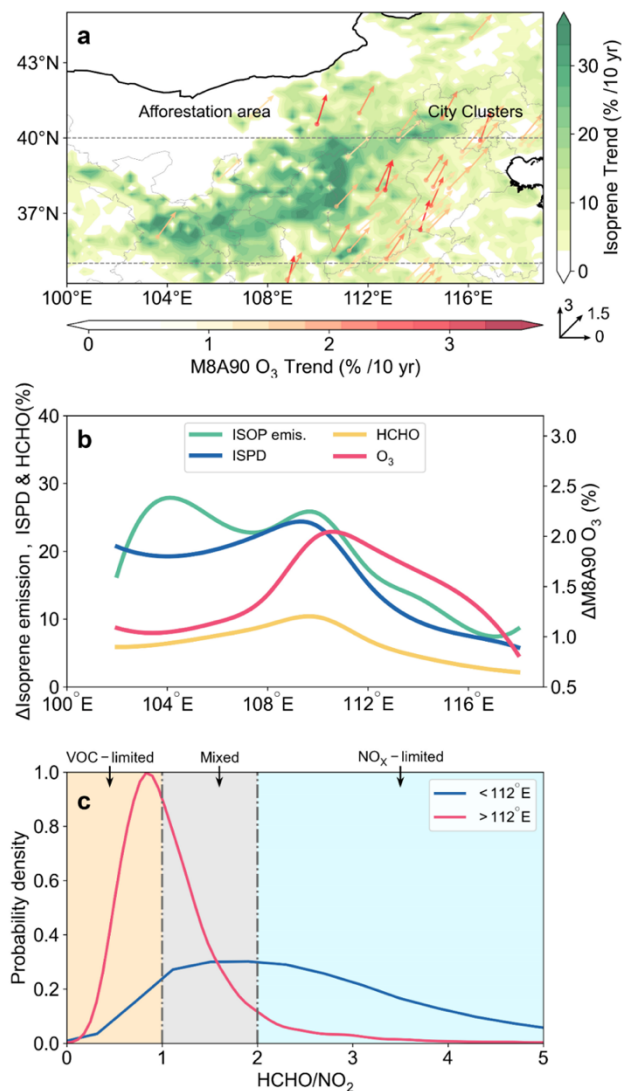


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305 **Figure 1. Changes of leaf area index (LAI) and ozone in Northern China.**  
 306 (a) LAI changes in summer between 2019 and 2001 (2019 minus 2001); pie  
 307 pie chart shows the frequency of LAI changes in a; the grey rectangle shows  
 308 regions in b. (b) Surface  $O_3$  changes from 2013 to 2019 (2019 minus 2013,  
 309 marked with filled circles), with the green background representing the LAI in a.  
 310 Areas of the Loess Plateau and city clusters are indicated by red box and blue  
 311 lines. (c) Regional mean concentrations (bold lines and circles) and their trends  
 312 (thin lines) of summer LAI (green), tropospheric column ozone (blue) and  
 313 surface ozone (red) over the Loess Plateau. Shadows show the 25<sup>th</sup> and 75<sup>th</sup>  
 314 percentiles, respectively.

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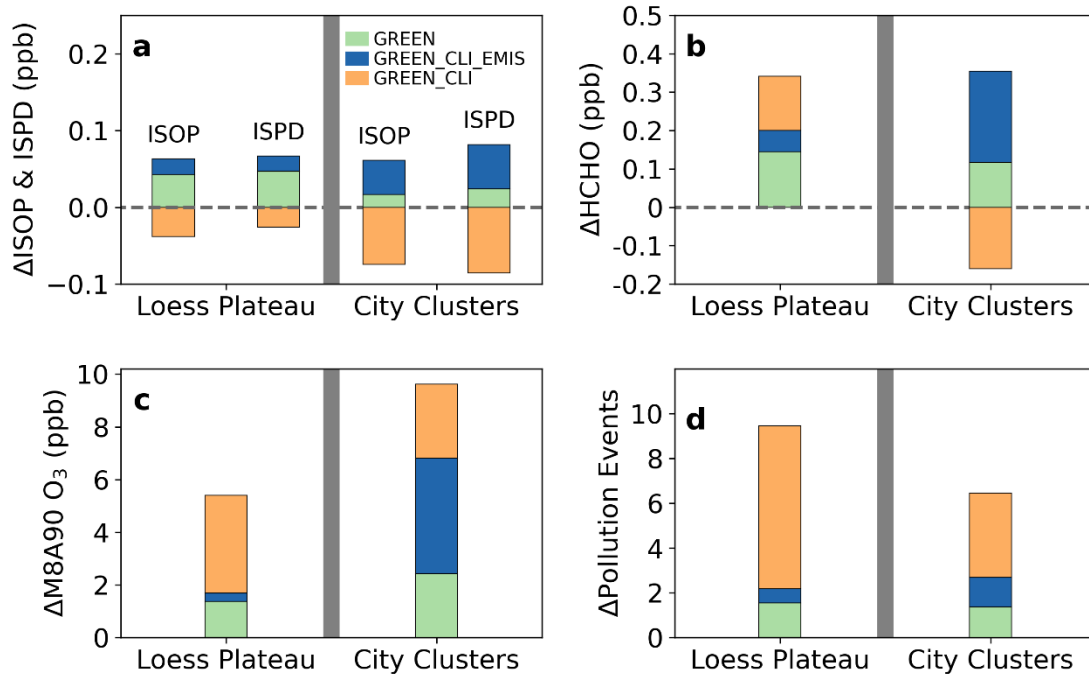
318 **Figure 2. Effect of afforestation on O<sub>3</sub> chemistry in northern China.**

319 (a) Simulation of isoprene emission (green contours) and M8A90 O<sub>3</sub> (red  
 320 arrows) trends from 2001 to 2019. Both directions and colors of the arrows  
 321 indicate the ozone change rates. Only M8A90 O<sub>3</sub> trends in high-NO<sub>x</sub>-emission  
 322 areas are plotted (Areas with exceedance of 90<sup>th</sup> percentiles of summertime  
 323 NO<sub>x</sub> emissions in this domain). (b) Meridional population-weighted mean (35–  
 324 41°N, area between dashed grey lines in a) changes of isoprene emission,  
 325 ISPD (lumped methacrolein, methyl vinyl ketone, methyl glyoxal, etc), HCHO  
 326 and M8A90 O<sub>3</sub> between the periods of 2001-2003 and 2017-2019 (2017-2019  
 327 minus 2001-2003). (c) Ratios of HCHO to NO<sub>2</sub> based on GOME-2 satellite data  
 328 in two regions. Note that meteorology and anthropogenic emissions were fixed  
 329 in 2010 while land cover/land change data was updated each year.

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**Figure 3. Effect of climate change to emission and chemistry.**

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Contribution of greening (green bar), climate change (enhanced-biogenic-

336

emission effect (blue bar) and accelerated-reaction-rates effect (orange bar))

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to isoprene and ISPD (a), HCHO (b), M8A90 O<sub>3</sub> (c), and pollution events (d)

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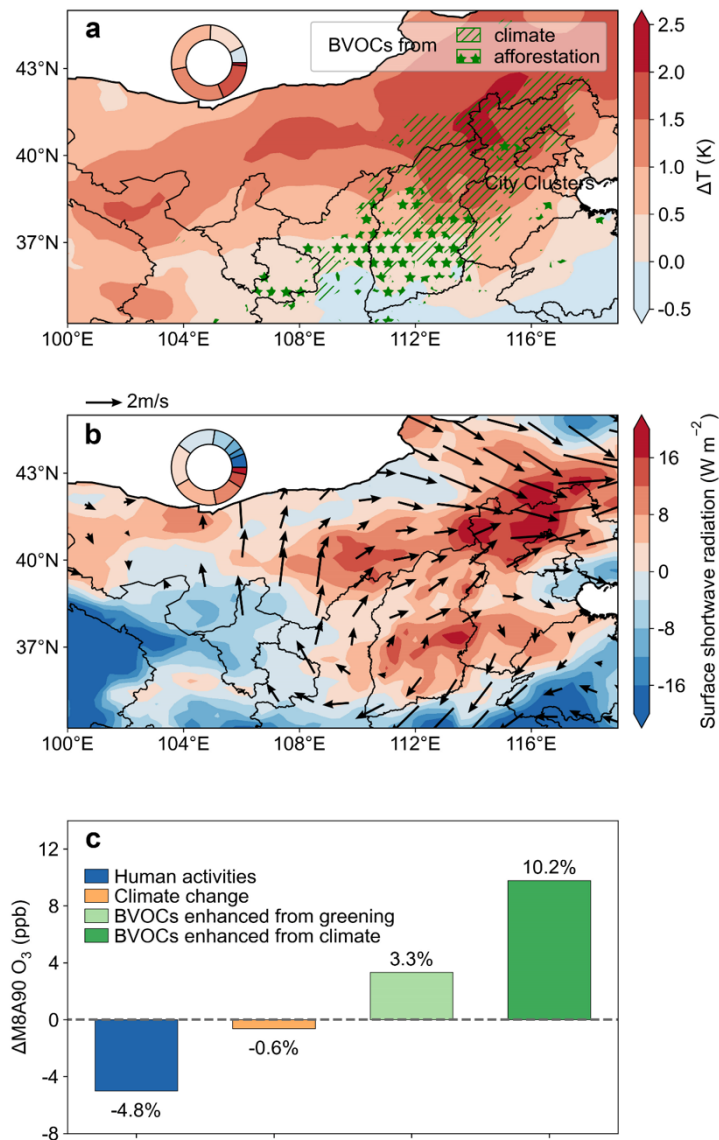
between 2001-2003 and 2017-2019 (2017-2019 minus 2001-2003). The

339

averaged areas are shown in Figure. 1b.

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**Figure 4. Impacts of climate change and vegetation greening on O<sub>3</sub> pollution in northern China.**

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(a) Anomalous 2 m temperature during extreme pollution events between 2020 and 2050. The regions with an exceedance of

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30% increment in isoprene emissions are illustrated by shadow.

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(b) Anomalous surface shortwave radiation and anomalous flows in 850hPa between 2020 and

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2050.

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(c) Effects of human activities, climate and greenness to future O<sub>3</sub> in city clusters. Days with an exceedance of 90<sup>th</sup> percentiles across City Clusters in

350

M8A90 O<sub>3</sub> are defined as extreme pollution events. Note: Pie charts in a and b

351

represent the frequency of changes in temperature and shortwave radiation,

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respectively; all differences are calculated between two periods of 2049-2051

353

and 2019-2021 (2050-2020).

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# EXPERIMENTAL PROCEDURES

## Resource availability

### Lead contact

Further information and requests for resources and reagents should be directed to and will be fulfilled by the lead contact, Aijun Ding ([dingaj@nju.edu.cn](mailto:dingaj@nju.edu.cn)).

### Materials availability

This study did not generate new unique materials.

### Data and code availability

MODIS Satellite datasets are available from EARTHDATA (<https://earthdata.nasa.gov/>). NO<sub>2</sub> column datasets are available from OMI (<https://www.temis.nl/index.php>). O<sub>3</sub> column datasets are available from OMI ([https://acd-ext.gsfc.nasa.gov/Data\\_services/cloud\\_slice/new\\_data.html](https://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/new_data.html)). The CMIP5 RCP8.5 data are available from NCAR (<https://rda.ucar.edu/datasets/ds316.1>). The ozone data set used in this study can be downloaded from the website (<https://doi.org/10.6084/m9.figshare.21158743.v1>).

### **Observational and reanalysis data.**

Summer ground-based observations of O<sub>3</sub>, nitrogen dioxide (NO<sub>2</sub>) and CO concentrations at more than 1400 stations since 2013 are obtained for model validation. The monitoring data are archived at the air monitoring data center of the Ministry of Ecology and Environment (MEE) of China. In 2013 MEE established the monitoring sites mainly in capital city of each province and then enlarged sites to most prefectural-level cities in China. Here, we evaluate modeling results with observational pollutants in northern China (Figure S4). Apart from surface observational data, column O<sub>3</sub> datasets from OMI are also used.

### **Regional chemical transport model.**

Weather Research and Forecast – Community Multiscale Air Quality (WRF-CMAQ) modeling system was employed to investigate O<sub>3</sub> changes in northern China. This modelling system considers complex photochemical reactions and is widely used to understand the impacts on air pollutants (especially for O<sub>3</sub>) from meteorology and emission changes<sup>61-64</sup>.

In this study, the model domain covered China and its surrounding area, centered at 39°N, 106.8°E with a horizontal grid resolution of 27 km, and 27 vertical layers from surface to 100 hPa. NCEP global final analysis (FNL) data were used as the initial and lateral boundary conditions of meteorological

variables with a  $1^{\circ}\times 1^{\circ}$  spatial resolution that updates every 6 hours. While for modeling atmospheric chemistry, CMAQv5.1 was used and the major gas phase chemistry was represented by Carbon Bond version 05 (CB05) combined with Aerosol Module version 6 (AERO6). The initial and boundary conditions of chemical compositions came from a previous study (multi-year averaged FSDSMAM-hist using Community Earth System Model )<sup>65</sup> and, subsequently, dynamical downscaling to improve regional model simulations<sup>66,67</sup>. The key configuration of WRF-CMAQ included the Rapid Radiative Transfer Model (RRTM) for longwave and shortwave radiation, the Noah Land Surface Model for land-atmospheric interactions, the Kain-Fritsch scheme for cumulus parameterization, the Lin microphysics scheme and the ACM2 boundary layer scheme. The anthropogenic emissions of China were obtained from Multi-resolution Emission Inventory for China (MEIC), which were developed by Tsinghua University and updated to 2017 levels<sup>68</sup>.

Nine simulation scenarios were performed to investigate the effects of BVOC emissions (Supplementary Table S1).

- (1) One BASE scenario was designed for model evaluation;
- (2) Three HISTORY scenarios (GREEN, GREEN\_CLI\_EMIS and GREEN\_CLI) were designed to estimate the impacts of afforestation projects and climate change on O<sub>3</sub> concentrations. For the GREEN scenario, only vegetation cover in MEGAN changed annually, leading to changes in BVOC emissions, while the meteorological parameters and anthropogenic emissions in CMAQ were fixed

in 2010. The GREEN\_CLI\_EMIS scenario was the same as GREEN, but meteorological parameters and land cover drive MEGAN vary yearly. The GREEN\_CLI scenario was the same as GREEN\_CLI\_EMIS, but the meteorological parameters also vary yearly in CMAQ. Thus, the GREEN scenario represents the impact of afforestation projects on O<sub>3</sub> concentrations by changing BVOC emissions; the GREEN\_CLI\_EMIS scenario represents the impact of BVOC emissions (driven by climate change and afforestation projects) on O<sub>3</sub> concentrations; and the GREEN\_CLI scenario examines the overall effects of climate change and afforestation projects on O<sub>3</sub> pollution.

(3) Five FUTURE scenarios have similar simulation designs to isolate the impacts of anthropogenic activities, afforestation projects, and climate change on future O<sub>3</sub> pollution. More specific details of our experimental designs can be found in supplemental information Experimental design.

### **Biogenic emissions.**

The calculations of biogenic emissions in this study were performed using Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGANv2.1)<sup>69</sup>. It is widely used in simulation of BVOCs in China<sup>22, 24, 50, 70</sup>. MEGANv2.1 calculates emissions for 19 emission species include isoprene and monoterpenes based on following algorithm:

$$F_i = \gamma_i \sum \epsilon_{i,j} \chi_j \quad (1)$$

where  $F_i$ ,  $\varepsilon_{ij}$  and  $\chi_j$  are emission amount, standard emission factor and fractional coverage of each plant functional type  $j$  of chemical species  $i$ .  $\gamma_j$  is emission activity factor, which is defined based on canopy environment coefficient, leaf area index (LAI), light ( $\gamma_L$ ), temperature ( $\gamma_T$ ), leaf age ( $\gamma_{LA}$ ), soil moisture ( $\gamma_{SM}$ ), and CO<sub>2</sub> inhibition ( $\gamma_{Cl}$ ):

$$\gamma_i = C_{CE} LAI \gamma_{L,i} \gamma_{T,i} \gamma_{LA,i} \gamma_{SM,i} \gamma_{Cl,i} \quad (2)$$

Here, PFT data was from MODIS MCD12Q1 datasets and was classified from 8 vegetation types to 16 PFT types in MEGANv2.1 according to climatic criteria defined in previous study<sup>71</sup>. LAI data was also from MODIS datasets, with MODIS MOD15A2H for 2001 and MCD15A2H for 2002-2019, respectively. The PFT product is generated each year while the LAI products are composited every 8 days<sup>31,72</sup>. At last, meteorological data was from WRF simulation.

## **Socioeconomic and climate scenarios**

To quantify the relative impacts of future greening, climate change and anthropogenic emission change on O<sub>3</sub> concentrations, we vary these factors one at a time and compare the M8A90 O<sub>3</sub> attributable to each of these factors independently between 2020s and 2050s using the SSP-RCP scenarios.

In this paper, the future human activities scenarios are from SSP-RCPs of Coupled Model Intercomparison Project Phase 6 (CMIP6), which are combined with two dimensions<sup>73</sup>. One dimension is Shared Socioeconomic Pathways

(SSPs). The SSPs describe future changes in the energy mix, technological progress, population growth, diets, global collaboration, and so on. For example, SSP5 describes a high fossil-fueled development world<sup>51,74</sup>. The other dimension is Representative Concentration Pathways (RCPs), which are the outcomes of climate policies. RCP8.5 assume paths limiting radiative forcing to  $8.5 \text{ W m}^{-2}$  in 2100. The CMIP5 RCP8.5 data are downloaded from NCAR<sup>75</sup>.

In this study, the anthropogenic emissions (e.g.,  $\text{NO}_x$  and VOCs emissions) are following SSP5-8.5 scenario<sup>74</sup>. Although cleaner combustion technologies and the shifting from coal use to liquefied petroleum gas (LPG) and electricity for energy, industry, and residential sectors substantially decrease ozone precursor gases emissions. Transportation, however, will primarily increase anthropogenic emissions in city clusters in 2050, associated with the growing economy and increasing car ownership in China<sup>76,77</sup>. The results start from 2015, and emissions are provided every 10 years during 2020 to 2100. Therefore, the  $\text{NO}_x$  and VOC emissions in different years are scaled up or down based on MEIC emission inventory in 2015.



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## **AUTHOR CONTRIBUTIONS**

A.D. and X. H. designed the research. J.X. performed the simulations. J.X., S. L., X. H, and A. D. performed the overall analysis. J.X., and S. L. drafted the paper with the assistance from all authors.

## **COMPETING INTERESTS**

The authors declare no competing interests.

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