

1 **Dissimilar Roles of Aerosols, Nitrogen Deposition and Ozone on the Terres-**
2 **trial Carbon Sink in China during 2010-2020**

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18
19 **Abstract**

20 China's Clean Air Action (CAA) plan implemented since 2013 has significantly altered atmos-
21 pheric composition, and yet its impact on the terrestrial carbon sink remains unclear. This study
22 employed the Regional Earth System Model (RegESM), an online-coupled climate–chemistry–
23 ecosystem modeling framework, to quantify the impacts of aerosols, surface ozone (O₃), and
24 nitrogen deposition on China's net ecosystem productivity (NEP) from 2010 to 2020. The re-
25 sults show that aerosols enhanced China's NEP by 17.93 TgC yr⁻¹ (4.49% of the total NEP),
26 primarily by increasing diffuse radiation, with the most pronounced effects in Southern and
27 Eastern China. Nitrogen deposition further increased NEP by 37.98 TgC yr⁻¹ (9.52%),

28 concentrated in Central and Southern regions. In contrast, O₃ pollution reduced NEP by 51.33
29 TgC yr⁻¹ (12.90%), particularly in the forest-dominated Southeast. The positive impacts of aer-
30 osols and nitrogen deposition on the carbon sink weakened over time, whereas the negative
31 influence of O₃ was increasing. The combined effects indicate that CAA-induced atmospheric
32 chemistry changes led to a shift in the dominant atmospheric drivers of China's terrestrial car-
33 bon sink, from enhancement by aerosols and nitrogen deposition to suppression by ozone. Our
34 findings highlight the need for stronger O₃ pollution control to achieve co-benefits between air-
35 quality improvement and carbon neutrality.

36 **1 Introduction**

37 Terrestrial ecosystems act as major carbon sinks, sequestering atmospheric carbon dioxide
38 (CO₂) through plant photosynthesis, and constitute a fundamental natural process for mitigating
39 global climate change (Friedlingstein et al., 2023; Piao et al., 2013; Yuan et al., 2025). Under
40 ongoing global warming, the dynamics of carbon sinks are regulated not only by climatic fac-
41 tors such as temperature and precipitation (Cao et al., 2023; Post et al., 2018; Ren et al., 2020),
42 but also by variations in atmospheric composition (Zhou et al., 2021). Among these, aerosols,
43 O₃, and atmospheric nitrogen deposition have been identified as key atmospheric pollutants
44 affecting terrestrial carbon sequestration (Liu et al., 2022; Zhou et al., 2024). As a crucial com-
45 ponent of the global carbon cycle, terrestrial ecosystems in China sequester approximately
46 0.20–0.25 PgC yr⁻¹, playing an essential role in supporting the achievement of the national car-
47 bon neutrality target (Piao et al., 2022; Xia et al., 2025; Yue et al., 2021). Therefore, assessing
48 the responses of carbon sinks to multiple atmospheric composition changes is of great scientific
49 significance for understanding both the global carbon cycle and climate feedback mechanisms.

50 Aerosols influence vegetation photosynthesis and carbon sequestration primarily through
51 radiative forcing (Shu et al., 2022; Zhou et al., 2022). Aerosol scattering and absorption reduce
52 surface solar radiation and can suppress vegetation photosynthesis (Doughty et al., 2010;
53 Kuniyal and Guleria, 2019). In the meantime, enhanced diffuse radiation increases light use
54 efficiency of plants, leading to the diffuse fertilization effect (Gu et al., 2003; Mercado et al.,
55 2009). Aerosols also influence cloud microphysics by modifying droplet formation and lifetime,

56 which further affects regional precipitation and water availability for vegetation (Li et al., 2020;
57 Unger et al., 2017). Consequently, the net effect of aerosols on photosynthesis exhibits marked
58 spatial heterogeneity, with both enhancement and suppression reported in highly polluted re-
59 gions such as eastern China (Strada and Unger, 2016; Wang et al., 2018; Xie et al., 2020).

60 In addition, near-surface O₃ impairs plant carbon uptake through direct physiological
61 damage (Lei et al., 2022; Unger et al., 2020). O₃ enters leaves through stomata and induces
62 reactive oxygen species at the cellular level, leading to degradation of photosynthetic pigments,
63 suppressed Rubisco activity, premature leaf senescence, and defoliation, all of which inhibit
64 photosynthetic carbon assimilation (Wittig et al., 2007). Evidence from O₃-FACE (free-air O₃
65 concentration enrichment) experiments shows that a 10 ppb increase in O₃ concentration can
66 lower crop productivity by 5–15% (Feng et al., 2015). In China, summertime O₃ peaks often
67 coincide with the peak growing season of vegetation, particularly in the North China Plain and
68 the Yangtze River Delta, posing a notable threat to regional carbon sequestration (Lei et al.,
69 2022; Li et al., 2024; Yue et al., 2017).

70 Furthermore, atmospheric nitrogen deposition is a major external nitrogen source for ter-
71 restrial ecosystems and exerts both positive and negative effects on carbon sinks (Chen et al.,
72 2015; Lu et al., 2021). In nitrogen-limited systems, such as temperate forests and grasslands,
73 moderate deposition can enhance photosynthesis and biomass accumulation, thereby increas-
74 ing carbon sequestration (Cen et al., 2025; Lu et al., 2016; Peng et al., 2025). When inputs
75 become excessive, however, they can induce soil acidification, biodiversity loss, and broader
76 ecosystem degradation, a condition known as nitrogen saturation (Chen et al., 2015; Yue et al.,
77 2016). China receives some of the highest nitrogen deposition levels globally, with annual av-
78 erages of 15–20 kg N ha⁻¹ yr⁻¹ and hotspots surpassing 30 kg N ha⁻¹ yr⁻¹, raising increasing
79 concerns about long-term ecological impacts (Liu et al., 2022; Liu et al., 2013; Yu et al., 2019).

80 China has long faced the dual pressures of severe air pollution and growing greenhouse
81 gas emissions (Tu et al., 2019; Wang et al., 2024). Rapid economic expansion in the early 2010s
82 was accompanied by persistent increases in fine particulate matter (PM_{2.5}) concentrations (Hao
83 et al., 2020). Since 2013, successive Clean Air Action Plans have led to a substantial decline in
84 PM_{2.5} levels (Xue et al., 2019; Yue et al., 2020; Zheng et al., 2018). At the same time, near-

85 surface summertime O₃ has risen sharply (Liu et al., 2018; Zhou et al., 2024), while nitrogen
86 deposition has slowed in growth but remains at a high level (Liu et al., 2024). These changes
87 not only reflect the outcomes of emission control policies but also reshape the regional atmos-
88 pheric chemical environment, potentially exerting complex and combined effects on carbon
89 sinks (Liu et al., 2022; Zhou et al., 2024). The rapid transition in atmospheric composition
90 during 2010–2020 provides an unprecedented large-scale natural experiment for disentangling
91 the relative roles of aerosols, O₃, and nitrogen deposition in altering China’s carbon sink. How-
92 ever, most existing studies have examined these drivers in isolation, relied on offline or statis-
93 tical frameworks that cannot capture dynamic climate–chemistry–ecosystem feedbacks, and
94 rarely compared responses across ecological regions (Unger et al., 2020; Yue et al., 2017; Zhou
95 et al., 2024).

96 Here, we employ an improved regional climate–chemistry–ecosystem online-coupling
97 model, RegESM (Xie et al., 2024; Zhang et al., 2025), to quantify the impacts of aerosols, O₃,
98 and nitrogen deposition on China’s terrestrial carbon sinks during 2010–2020. RegESM incor-
99 porates two-way interactions among climate, atmospheric chemistry, and biogeochemical pro-
100 cesses and has been extensively evaluated over East Asia (Ma et al., 2023; Xie et al., 2024; Xie
101 et al., 2020; Zhang et al., 2025). Our objective is to isolate the contributions of individual at-
102 mospheric components to changes in China’s carbon sinks based on RegESM after its assess-
103 ment using multiple observational datasets. These results offer new insight into the ecological
104 consequences of rapid atmospheric composition changes and provide a scientific foundation
105 for coordinated multi-pollutant control and ecosystem management under China’s carbon-neu-
106 trality goals.

107 **2 Data and Methods**

108 **2.1 The RegESM model**

109 In this study, we employed the RegESM, an improved extension of the RegCM-Chem-
110 YIBs regional climate–chemistry–ecosystem modeling framework (Xie et al., 2024; Xie et al.,
111 2019; Zhang et al., 2025). The original RegCM-Chem-YIBs couples the RegCM4 regional

112 climate model (Giorgi et al., 2012), the radiative interactive gas-phase chemistry module Chem
113 (Shalaby et al., 2012), and the YIBs terrestrial ecosystem model (Yue and Unger, 2015) to
114 represent interactive processes among atmospheric dynamics, chemistry, and terrestrial carbon
115 cycles (Xie et al., 2024). Building upon this foundation, RegESM strengthens two-way feed-
116 back among the atmosphere, atmospheric chemistry, and land surface processes, enabling a
117 more realistic simulation of biogeochemical cycles (Zhang et al., 2025). The enhanced coupling
118 allows land surface changes, such as vegetation dynamics and soil moisture variations, to more
119 directly influence atmospheric composition, radiation, and meteorology, while atmospheric
120 and chemical variations simultaneously affect ecosystem processes (Xie et al., 2024; Zhang et
121 al., 2025). This bidirectional integration improves the model's capability to capture transient
122 and spatially heterogeneous climate–ecosystem–chemistry interactions, which are crucial for
123 regional climate change and carbon budget assessments (Zhang et al., 2025).

124 The RegESM framework used in this study integrates RegCM4 as the dynamical core for
125 simulating regional climate processes at a high resolution, the Chem module for interactive
126 gas-phase and aerosol chemistry coupled with radiation and meteorology, and the YIBs land
127 surface model for calculating biophysical processes such as photosynthesis, transpiration, and
128 energy balance, along with biogeochemical cycles of carbon and nitrogen (Giorgi et al., 2012;
129 Shalaby et al., 2012; Xie et al., 2024; Yue and Unger, 2015). In RegESM, the influence of
130 atmospheric nitrogen deposition on terrestrial carbon fluxes is represented through the online
131 coupling between the chemistry and land components. Atmospheric nitrogen deposition is cal-
132 culated online by the chemistry component as dry and wet deposition fluxes of reduced and
133 oxidized nitrogen (NH_x and NO_y), which are then passed to the land component as external
134 nitrogen inputs. These inputs affect soil inorganic nitrogen availability and subsequently influ-
135 ence plant productivity, ecosystem respiration, and net ecosystem productivity. Therefore, the
136 effect of nitrogen deposition on carbon fluxes is represented as the integrated result of nitrogen
137 input and land biogeochemical processes, rather than as a simple linear fertilization effect.
138 These components are linked through an improved coupling mechanism that ensures the con-
139 sistent exchange of meteorological, chemical, and biogeophysical variables at each model
140 timestep, enabling fully interactive simulations in which land, atmosphere, and chemistry

141 evolve in a physically coherent manner (Xie et al., 2024; Zhang et al., 2025). This model has
 142 been widely applied in East Asia (Xie et al., 2025; Xie et al., 2019; Zhang et al., 2025; Zhang
 143 et al., 2024).

144 We used net ecosystem productivity (NEP) as an indicator for characterizing carbon
 145 sources and sinks ($NEP > 0$ suggests a carbon sink). NEP was calculated as the difference
 146 between gross primary production (GPP) and the sum of autotrophic respiration (Ra) and het-
 147 erotrophic respiration (Rh) (Xie et al., 2025; Yue et al., 2021). It is noteworthy that the NEP
 148 estimated in this study does not account for lateral carbon transfers.

149 2.2 Ozone Damage Scheme

150 Once surface O_3 enters plants through the stomata, it directly damages plant cellular struc-
 151 tures and suppresses the photosynthetic rate, thereby reducing vegetation productivity. In the
 152 YIBs vegetation module of the RegESM model, a semi-mechanistic parameterization scheme
 153 is employed to represent the impacts of O_3 on plants (Sitch et al., 2007; Yue and Unger, 2015):

$$154 \quad B = B_{tot} \cdot K, \quad (1)$$

155 where B denotes the photosynthetic rate under O_3 exposure, B_{tot} represents the total leaf pho-
 156 tosynthetic rate, and K is the remaining proportion of photosynthetic capacity after O_3 stress.
 157 This proportion is determined by the stomatal O_3 flux that exceeds a specified threshold:

$$158 \quad K = 1 - b \cdot \max[(K_{ozn} - K_{ozncrit}), 0], \quad (2)$$

159 where b denotes the vegetation sensitivity parameter to O_3 derived from observational data.
 160 $K_{ozncrit}$ represents the threshold of O_3 -induced damage to vegetation, and K_{ozn} denotes the
 161 O_3 flux entering the leaf through stomata:

$$162 \quad K_{ozn} = \frac{[O_3]}{r_b + \frac{\kappa_{O_3}}{r_s}}, \quad (3)$$

163 where $[O_3]$ denotes the O_3 concentration at the canopy top, r_b is the boundary layer resistance,
 164 κ_{O_3} is the ratio of O_3 leaf resistance to water vapor blade resistance, and r_s is the stomatal
 165 resistance accounting for the effects of O_3 :

$$166 \quad r_s = g_s \cdot K. \quad (4)$$

167 g_s denotes the leaf conductance unaffected by O_3 exposure. By simultaneously solving

168 Equations (2), (3), and (4), a quadratic term with respect to K is obtained, which can be solved
169 analytically.

170 2.3 Experimental design and input data

171 The simulation domain covers most of East Asia (Fig. S1), centered at 36° N and 107° E.
172 The horizontal resolution is 30 km, with 18 vertical layers. To quantify the independent con-
173 tributions of aerosol, O₃ damage, and atmospheric nitrogen deposition to China’s terrestrial
174 carbon sink during 2010–2020, four sensitivity experiments were conducted (Table 1): a base-
175 line simulation without these effects (Base), and three single-factor cases that enabled only
176 aerosol (Ctrl_AOD), O₃-induced vegetation damage (Ctrl_O₃), and nitrogen deposition im-
177 pacts (Ctrl_Ndep). In the Ctrl_AOD experiment, aerosols were fully coupled to meteorology,
178 so that the direct aerosol radiative effect and the associated meteorological responses were
179 represented in the simulations, while aerosol indirect effects through cloud processes were not
180 included. The difference between each sensitivity case and the Base run represents the corre-
181 sponding individual effect. All simulations were preceded by a one-year spin-up to reduce the
182 influence of initial conditions. To further assess regional responses, China was divided into six
183 representative subregions (Fig. S2), and statistical analyses were performed for each.

184

185 **Table 1.** Numerical model experiments.

| Simulations | Periods | Aerosol direct radiative effect | O ₃ damage | Atmospheric nitrogen deposition |
|---------------------|-----------|---------------------------------|-----------------------|---------------------------------|
| Base | 2010-2020 | off | off | off |
| Ctrl_AOD | 2010-2020 | on | off | off |
| Ctrl_O ₃ | 2010-2020 | off | on | off |
| Ctrl_Ndep | 2010-2020 | off | off | on |

186

187 The initial and boundary meteorological fields were taken from the ECMWF (European
188 Centre for Medium-Range Weather Forecasts) ERA-Interim reanalysis with a temporal

189 resolution of 6 h and a horizontal resolution of $1.5^\circ \times 1.5^\circ$ (Hersbach et al., 2020). Aerosol
190 initial and boundary conditions were provided by the global chemical transport model (MO-
191 ZART) (Emmons et al., 2010; Horowitz et al., 2003). Background CO₂ fields were constrained
192 by three-dimensional concentrations from NOAA CarbonTracker (CT) reanalysis (Peters et al.,
193 2007). The initial parameters for the YIBs model were derived from soil carbon stocks based
194 on equilibrium tree height and a 30-year harvest cycle (Yue and Unger, 2015). Vegetation
195 cover was prescribed from MODIS and AVHRR (Advanced Very High Resolution Radiometer)
196 datasets (Lawrence and Chase, 2007). Anthropogenic emissions in China were taken from the
197 Multi-resolution Emission Inventory for China (MEIC) (Geng et al., 2024; Li et al., 2017;
198 Zheng et al., 2018).

199 **2.4 Validation data**

200 We employed monthly mean aerosol optical depth (AOD) data from the MODIS sensor
201 onboard NASA's Terra satellite (MOD08_M3.061). The data have a spatial resolution of $1^\circ \times$
202 1° and are retrieved using three algorithms: the Dark Target, Deep Blue, and combined ap-
203 proaches (Levy et al., 2013). Ground-level O₃ observations were obtained from 366 monitoring
204 stations operated by the China National Environmental Monitoring Center (CNEMC). To eval-
205 uate the model's capability in simulating atmospheric nitrogen deposition, we employed pub-
206 licly available datasets (Liu et al., 2024; Zhu et al., 2025). These datasets integrate observations
207 with model outputs to provide nitrogen deposition estimates at both global and regional scales
208 over China. To assess the reliability of simulated CO₂, we used observations from the World
209 Data Centre for Greenhouse Gases (WDCGG). This dataset provides measured surface atmos-
210 pheric CO₂ concentrations and was used to evaluate the model's ability to reproduce observed
211 CO₂ levels. For the spatial distribution of CO₂, we additionally used CO₂ concentration fields
212 from CT (Peters et al., 2007). For GPP and net primary production (NPP) validation, we used
213 the global MODIS products MOD17A2H and MOD17A3H (Collection 6). The GPP data, at
214 8-day resolution, were derived using the radiation use efficiency algorithm, while NPP
215 (NPP=GPP-Ra) data were produced by annually accumulating GPP values, with a spatial res-
216 olution of 500 m (He et al., 2018; Madani et al., 2014).

217 2.5 Analytical Approach

218 Aerosol-induced meteorological changes are highly interdependent, making it challenging
219 to isolate their individual effects on terrestrial carbon cycling. To quantify the relative contri-
220 butions of these meteorological responses to vegetation carbon fluxes, we applied a multiple
221 linear regression framework. Standardized regression coefficients were used to assess the rel-
222 ative influence of each climate variable. This approach has been widely demonstrated as effec-
223 tive for disentangling the impacts of multiple environmental drivers on ecosystem processes
224 (Jung et al., 2017; Xie et al., 2025; Zhang et al., 2024).

225 The regression model is expressed as follows:

$$226 \Delta Y = A_1 \times \Delta X_1^{RadD} + A_2 \times \Delta X_2^{RadF} + A_3 \times \Delta X_3^{Temp} + A_4 \times \Delta X_4^{Precip} + A_5 \times \Delta X_5^{VPD} \\ 227 + \varepsilon \quad (5)$$

228 where ΔY denotes the difference in terrestrial carbon flux between the simulations Base and
229 Ctrl_AOD, respectively. ΔX_1^{RadD} , ΔX_2^{RadF} , ΔX_3^{Temp} , ΔX_4^{Precip} and ΔX_5^{VPD} denote
230 the differences in direct radiation, diffuse radiation, temperature, precipitation, and vapor pres-
231 sure deficit (VPD) between the simulations Base and Ctrl_AOD, respectively. A_i represents
232 the partial regression coefficient for different meteorological factors, indicating the sensitivity
233 of carbon flux to variations in these factors. ε is the residual term of the regression model. We
234 use the following equation to calculate the standardized regression coefficient B_i for compar-
235 ing the relative impacts of different meteorological factors:

$$236 B_i = A_i \times SD(\Delta X_i) \div SD(\Delta Y) \quad (6)$$

237 where $SD(\Delta X_i)$ and $SD(\Delta Y)$ represent the standard deviations of the changes in each mete-
238 orological factor and carbon flux, respectively. B_i quantifies the relative contribution of dif-
239 ferent meteorological factors to variations in carbon flux. This approach enables a quantitative
240 assessment of the individual impacts of changes in each meteorological factor induced by aer-
241 osol radiative effects on terrestrial carbon flux.

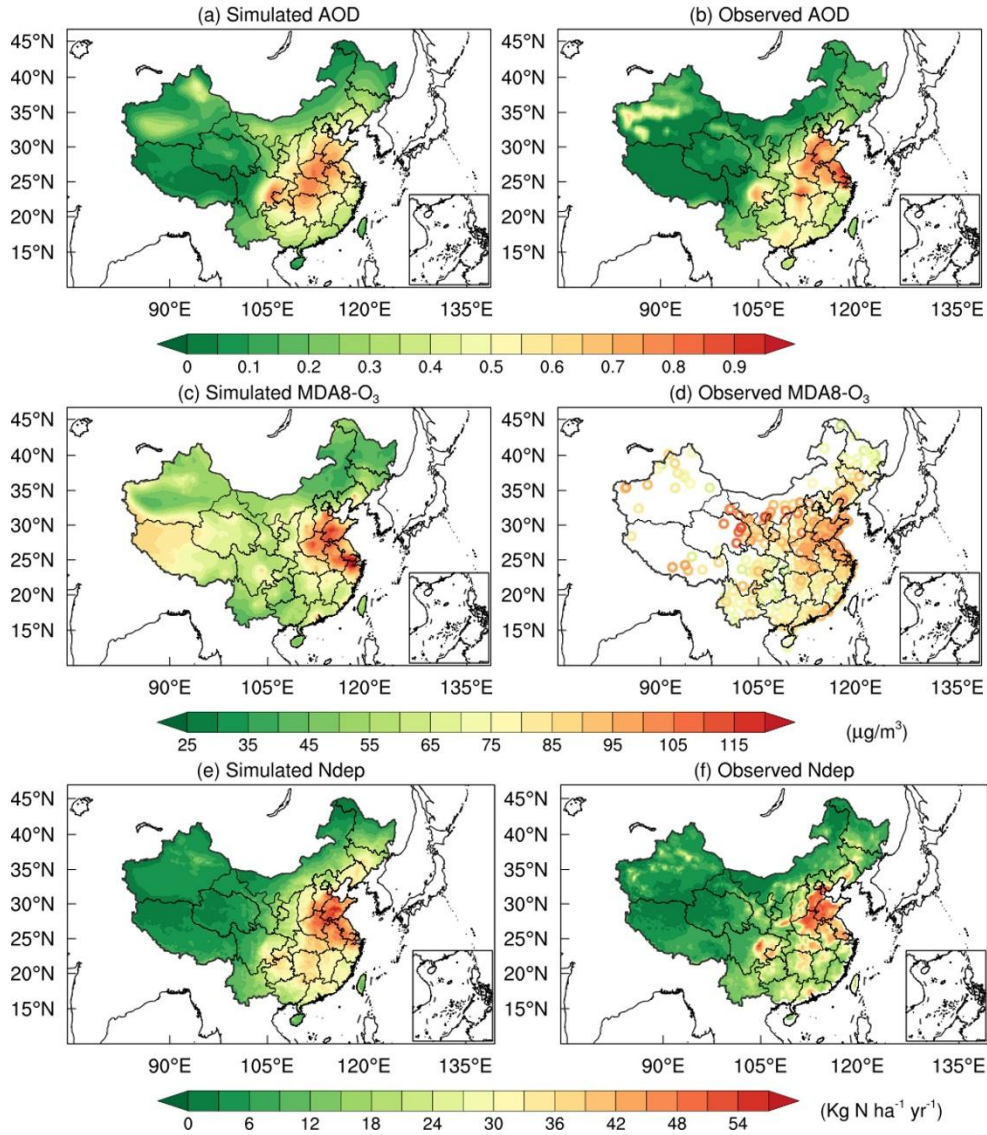
242 3 Results

243 3.1 Model validations

244 3.1.1 Aerosols, surface ozone, and atmospheric nitrogen deposition

245 We assessed the RegESM performance by comparing the 2010–2020 simulations with
246 multi-source observations. Simulated AOD showed good agreement with MODIS products in
247 both spatial distribution and magnitude (Fig. 1a, b). High AOD values are located over the
248 North China Plain and the Sichuan Basin, consistent with dense anthropogenic emissions in
249 these regions (Luo et al., 2014). The observations indicate that the national mean AOD de-
250 creased from 0.36 in 2010 to 0.28 in 2020, driven by air quality improvement policies. We
251 calculated statistical metrics, including the correlation coefficient (R), mean bias (MB), and
252 root mean square error (RMSE), to evaluate the model performance (Fig. S3). The RegESM
253 captures this trend with a correlation coefficient (R) of 0.71. However, compared with monthly
254 MODIS AOD, the model shows a minor underestimation (MB = -0.02), which can be at-
255 tributed primarily to uncertainties in the anthropogenic emission inventories (Xie et al., 2020).
256 Surface O₃ simulations reproduce both spatial patterns (Fig. 1c, d). The correlation with site
257 observations reaches 0.72 (Fig. S4). High concentrations in the North China Plain, the Yangtze
258 River Delta, and the Sichuan Basin are captured well, highlighting the model’s skill in simu-
259 lating O₃ fields. The simulated annual mean atmospheric nitrogen deposition flux ranges from
260 20 to 40 kg N ha⁻¹ yr⁻¹ in eastern agricultural and urban areas, consistent with reported values
261 of 25–35 kg N ha⁻¹ yr⁻¹ (Fig. 1e, f). The simulated national mean of 15.09 kg N ha⁻¹ yr⁻¹ is close
262 to the dataset range of 13.45–15.39 kg N ha⁻¹ yr⁻¹. The model also reproduces the observed
263 decline after the implementation of air pollution control policies in 2013, with a gradual de-
264 crease after 2015 (Fig. S5). These evaluations indicate that RegESM reliably simulates AOD,
265 O₃, and nitrogen deposition fields across China.

266



267

268 **Figure 1.** Annual mean AOD (a, b), maximum daily 8 h average (MDA8) O₃ (c, d), and At-
 269 mospheric nitrogen deposition (e, f) from model simulation (a, c, e) and observations (b, d, f).

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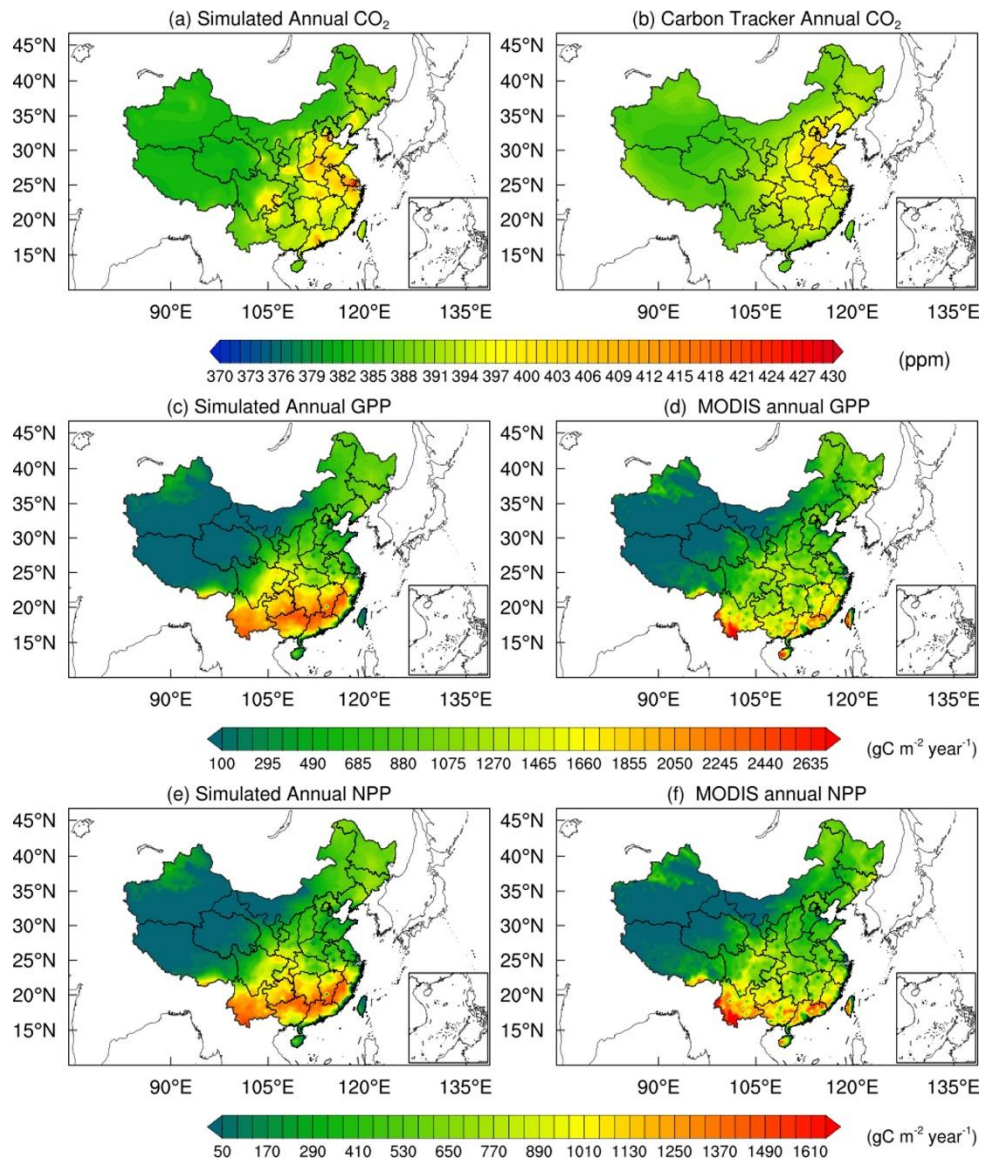
271 3.1.2 Atmospheric CO₂ concentrations, GPP, and NPP

272 Simulated CO₂ concentrations were compared with six stations from the WDCGG. The
 273 correlation coefficients range from 0.83 to 0.96 (Table S1). The YON site shows the best agree-
 274 ment (R = 0.96, MB = -1.10 ppm), likely due to minimal influence from terrestrial emissions.
 275 In contrast, HKG and HKO show larger biases, with overestimates of 3.1 ppm (R = 0.83) and
 276 3.3 ppm (R = 0.84), probably linked to unaccounted variability in urban sources in monthly

277 inventories. Nevertheless, the seasonal cycle is reproduced well at all sites (Fig. S6).

278 We further compared simulated CO₂ with the CarbonTracker CT2022 assimilation dataset
279 (Peters et al., 2007). The spatial correlation coefficient reaches 0.72 (Fig. 2a, b). High CO₂
280 concentrations appear over the Beijing–Tianjin–Hebei region, the Yangtze River Delta, the
281 Pearl River Delta, and the Sichuan Basin, consistent with intense industrial emissions. The
282 model slightly overestimates values in the Pearl River Delta, likely due to underrepresented
283 local sources and complex topography. Overall, RegESM effectively captures the spatial dis-
284 tribution of CO₂ concentrations.

285 Simulated GPP agrees well with MODIS in spatial distribution (Fig. 2c, d), with a spatial
286 correlation of 0.89. However, GPP from this study is larger than MODIS GPP by 7.4%, with
287 largest differences in Central (11.6%) and Southeast China (5.7%). Other studies also found
288 that MODIS GPP was underestimated at high values (Xie et al., 2019; Zhang et al., 2012). The
289 southeast-to-northwest decreasing gradient is reproduced, with high values over regions dom-
290 inated by forest ecosystems. The seasonal cycle of GPP is also captured (Fig. S7). The simu-
291 lated NPP exhibits a spatial distribution consistent with the MODIS (Fig. 2e, f), with a spatial
292 correlation coefficient of 0.86. Similar to GPP, the model overestimates NPP by 8.4%, mainly
293 due to the overestimation in Central (14.3%) and Northeastern (6.2%) China. These results
294 confirm the model’s ability to represent terrestrial carbon fluxes.



295
 296 **Figure 2.** Annual mean CO₂ (a, b), GPP (c, d), and NPP (e, f) from model simulation (a, c, e)
 297 and observations (b, d, f).
 298

299 3.2 Impacts of Aerosols on Meteorology and Carbon Sinks

300 3.2.1 Impacts of Aerosols on Meteorological Factors

301 During 2010–2020, the aerosol exerted a substantial influence on China’s surface radia-
 302 tion and near-surface climate (Fig. 3). Nationally, aerosol scattering reduced downward direct
 303 solar radiation by 8.81 W m⁻² and increased its diffuse component by 3.04 W m⁻², resulting in
 304 a net decrease of 5.77 W m⁻² in total downward shortwave radiation at the surface. Spatially,

305 these radiative changes were most pronounced over major urban agglomerations such as the
306 North China Plain, the Yangtze River Delta, and the Sichuan Basin, coinciding with regions of
307 high AOD associated with intensive anthropogenic emissions. These results are consistent with
308 previous modeling and satellite-based analyses (Wang et al., 2017; Xie et al., 2020), confirming
309 the robustness of the simulated radiative forcing patterns.

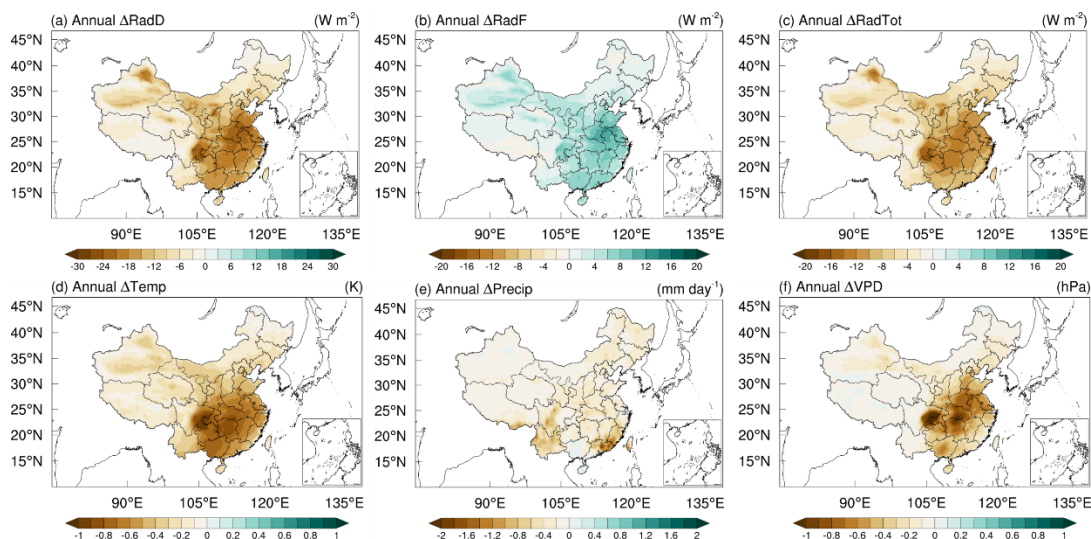
310 The reduction in surface solar radiation directly perturbed the regional energy balance and
311 atmospheric thermodynamics, resulting in a cooling effect over most of eastern and central
312 China. As shown in Fig. 3d, surface air temperature decreased significantly in the Sichuan Ba-
313 sin and coastal regions, with local maxima reaching -1.10 °C. In contrast, western and north-
314 eastern China experienced weaker changes, consistent with lower AOD levels. The simulated
315 national mean temperature decline of 0.32 °C agrees well with previous RegCM-based studies
316 (Wang et al., 2015; Xie et al., 2020). This widespread cooling is primarily attributed to aerosol-
317 induced dimming, which suppresses surface shortwave absorption and weakens boundary-
318 layer turbulence, thereby inhibiting vertical heat exchange and reducing near-surface tempera-
319 tures.

320 Aerosol also exerted a marked influence on regional hydrological processes. Precipitation
321 decreased across much of southern and southwestern China, with notable reductions in Guang-
322 dong, Fujian, Yunnan, and Sichuan provinces, where daily rainfall decreased by up to 2 mm
323 day^{-1} (Fig. 3e). On average, national precipitation declined by 0.23 mm day^{-1} . The reduction in
324 rainfall reflects the combined effects of radiative cooling and weakened convective activity.
325 Specifically, aerosol-induced surface dimming stabilizes the lower atmosphere and suppresses
326 the upward transport of moisture, while reduced latent heating further limits convective cloud
327 formation. These mechanisms together explain the widespread drying observed in the simula-
328 tions.

329 The VPD, a key indicator of plant water stress, also responded sensitively to aerosol forc-
330 ing. As shown in Fig. 3f, aerosols significantly reduced VPD over central and southeastern
331 China, with decreases of -0.30 to -0.60 hPa, and locally up to -1.20 hPa in Sichuan, Hebei, and
332 Jiangsu. The national mean reduction was -0.11 hPa. Lower VPD values imply a moister near-
333 surface environment and weaker atmospheric demand for evapotranspiration. Ecologically, this

334 alleviation of plant water stress can enhance stomatal conductance and facilitate photosynthetic
 335 carbon uptake, thereby partially compensating for the productivity loss caused by reduced solar
 336 radiation. Thus, the aerosol-induced decline in VPD represents an important indirect pathway
 337 through which aerosols modulate the terrestrial carbon cycle, linking atmospheric radiative
 338 forcing to ecosystem function.

339



340
 341 **Figure 3.** Annual mean changes in meteorological variables due to aerosol direct radiative
 342 effect during 2010–2020. (a) RadD, direct radiation; (b) RadF, diffuse radiation; (c) RadTot,
 343 total radiation; (d) Temp, air temperature; (e) Precip, precipitation; (f) VPD, vapor pressure
 344 deficit.

345

346 3.2.2 Effects of Aerosols on the Terrestrial Carbon Sink

347 During 2010–2020, the aerosol overall enhanced the productivity of China’s terrestrial
 348 ecosystems, increasing GPP and NEP by $293.28 \text{ TgC yr}^{-1}$ and $17.93 \text{ TgC yr}^{-1}$, accounting for
 349 3.98% and 4.49% of the national totals, respectively. Aerosols also increased Rh by 182.44
 350 TgC yr^{-1} over China, indicating that part of the aerosol-induced carbon sink enhancement was
 351 offset by enhanced soil carbon decomposition. Spatially, the responses of GPP and NEP to the
 352 aerosol radiative effect displayed significant heterogeneity, with the most pronounced enhance-
 353 ments occurring in South Central and East (Fig. 4a, b), where GPP increased by 0.32 gC m^{-2}
 354 day^{-1} and $0.31 \text{ gC m}^{-2} \text{ day}^{-1}$, respectively. These regions are characterized by dense forests and

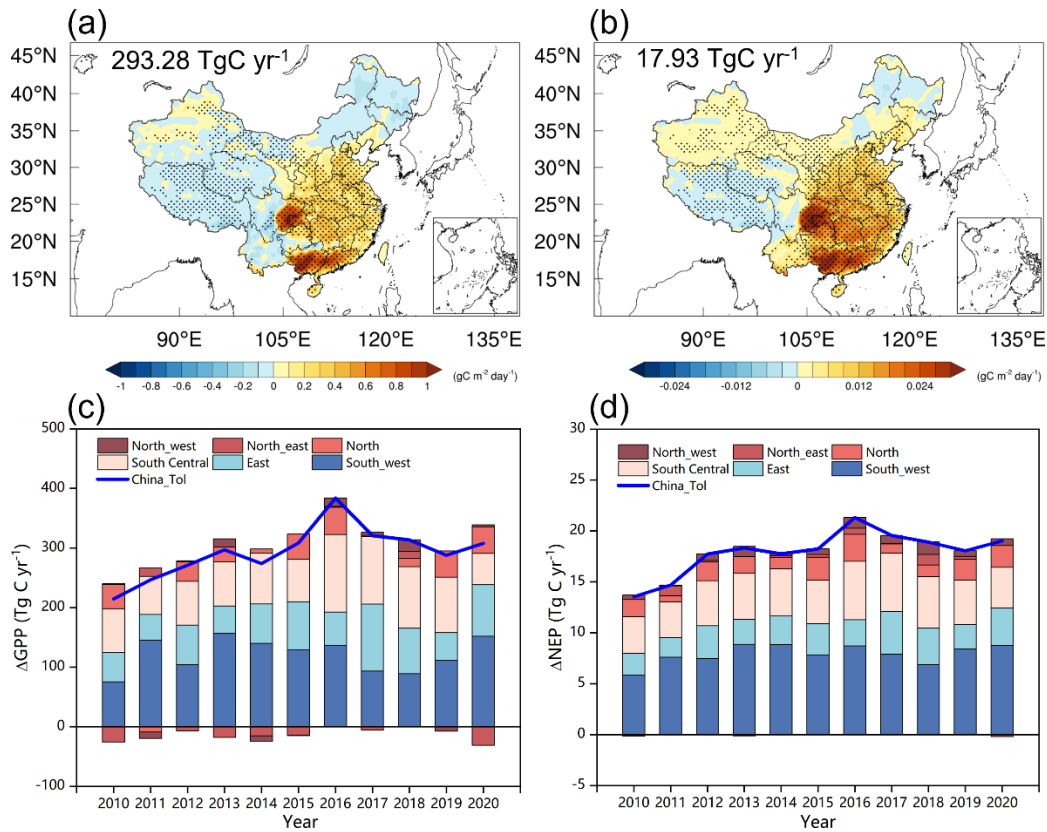
355 cropland ecosystems with high leaf area index, enabling them to fully exploit the additional
356 diffuse radiation induced by aerosols. Meanwhile, the high aerosol loading in these regions
357 ensured sufficient radiative perturbation, amplifying the improvement in canopy light-use ef-
358 ficiency. The spatial pattern of aerosol-induced Rh (Fig. S10a) further shows notable increases
359 in southern China, broadly consistent with the regions of enhanced ecosystem productivity,
360 suggesting that greater carbon input to soils likely stimulated microbial decomposition and
361 partially counteracted the NEP gain. In the Southwest, the response was more complex. Alt-
362 hough the mean GPP increased by $0.20 \text{ gC m}^{-2} \text{ day}^{-1}$, parts of Yunnan showed a negative effect.
363 This reduction likely results from excessive attenuation of solar radiation under the region's
364 unique topographic and climatic conditions, which constrained photosynthetic activity. Never-
365 theless, NEP in this region remained positive (approximately $0.01 \text{ gC m}^{-2} \text{ day}^{-1}$), suggesting
366 that the cooling effect of aerosols substantially suppressed ecosystem respiration, thereby com-
367 pensating for the reduced photosynthesis. In contrast, the North and Northwest exhibited weak
368 positive responses ($<0.07 \text{ gC m}^{-2} \text{ day}^{-1}$), while the Northeast showed slight inhibition (-0.04
369 $\text{gC m}^{-2} \text{ day}^{-1}$), probably due to aerosol-induced cooling delaying the onset of the growing sea-
370 son. Overall, the spatial patterns of GPP and NEP responses to the aerosol radiative effect show
371 a clear latitudinal gradient: the humid, high-biomass ecosystems in southern and eastern China
372 are most sensitive to diffuse radiation enhancement, whereas the high-latitude and arid regions
373 experience limited or even negative responses due to temperature and radiation constraints.

374 From 2010 to 2020, the influence of aerosols on carbon fluxes exhibited distinct interan-
375 nual variability (Fig. 4c, d). Both GPP and NEP showed an upward trend before 2016, with
376 GPP increasing from $214.66 \text{ TgC yr}^{-1}$ in 2010 to $384.00 \text{ TgC yr}^{-1}$ in 2016, and NEP rising from
377 $13.54 \text{ TgC yr}^{-1}$ to $21.31 \text{ TgC yr}^{-1}$. The synchronous growth of GPP and NEP indicates that the
378 aerosol radiative effect enhanced terrestrial carbon uptake mainly through photosynthetic ac-
379 tivity. The strong enhancement during 2015–2017 coincided with years of high aerosol loading
380 and a greater proportion of diffuse radiation, which improved canopy light-use efficiency under
381 humid and cloudy conditions. After 2018, the positive effect weakened slightly and stabilized
382 at a lower level. This reduction likely reflects the combined influence of cleaner atmospheric
383 conditions and changing meteorological patterns, including increased direct radiation and a

384 reduced diffuse fraction. Year-to-year variations were further modulated by hydroclimatic con-
385 ditions: higher humidity and cloud cover enhanced aerosol scattering efficiency, while drier or
386 cleaner years favored direct radiation and weakened the diffuse light advantage. Moreover, the
387 smaller NEP fluctuations compared to GPP imply a delayed response of ecosystem respiration,
388 as aerosol-induced cooling moderates' respiration more gradually than photosynthesis. Overall,
389 the interannual variability of GPP and NEP responses to the aerosol radiative effect highlights
390 the coupled influences of aerosol loading, radiation balance, and regional climate variability
391 on China's terrestrial carbon sink dynamics.

392 The effects of aerosols on GPP and NEP show pronounced seasonal variation (Fig. S8),
393 driven by the dynamic coupling between vegetation phenology and environmental factors. In
394 spring (March–May), aerosols increase GPP by 42.35 TgC (14.4%) and NEP by 2.82 TgC
395 (15.7%), making a notable contribution at the start of the growing season as rising temperatures
396 and rapid canopy expansion enhance diffuse radiation benefits, improving light-use efficiency;
397 meanwhile, moderate cooling suppresses respiration without causing thermal stress, further
398 boosting NEP. In summer (June–August), positive effects peak, with GPP rising by 173.62 TgC
399 (59.2%) and NEP by 10.15 TgC (56.6%); under high solar radiation and full canopy closure,
400 diffuse light penetration reaches its maximum, while cooling alleviates heat stress and reduces
401 respiration, driving NEP to its annual maximum. In autumn (September–November), aerosols
402 add 88.38 TgC to GPP (30.1%) and 3.95 TgC to NEP (22.0%), effectively extending the pho-
403 tosynthetic period as shorter days and reduced total radiation increase the proportion of diffuse
404 light, sustaining carbon storage. In winter (December–February), GPP declines slightly (-
405 11.07 TgC, -3.8%), but NEP shows a small positive gain (1.01 TgC, 5.6%) because cooling
406 strongly suppresses respiration, offsetting reduced photosynthesis. Overall, aerosol radiative
407 effects regulate seasonal carbon cycling by modifying radiation and thermal conditions. The
408 net impact depends on the trade-off between the fertilization effect of diffuse radiation and the
409 opposing effects of reduced total radiation and cooling. Summer emerges as the primary driver
410 of the annual net positive effect. Accurately quantifying this seasonal dynamic is crucial for
411 assessing the ecological and climatic consequences of anthropogenic aerosols.

412



413

414 **Figure 4.** Spatiotemporal variations in carbon flux changes caused by the aerosol radiative
 415 effect during 2010–2020. (a–b) Multi-year mean spatial patterns of GPP and NEP changes
 416 caused by the aerosol radiative effect. National totals are shown in each panel. Black dots de-
 417 note significant changes ($p < 0.01$). (c–d) Interannual variations of GPP and NEP changes
 418 caused by the aerosol radiative effect.

419

420 3.2.3 Contributions of Meteorological Factors to Carbon Sink Changes

421 We quantified the independent contributions of aerosol-induced meteorological changes
 422 to carbon fluxes using the multiple linear regression analysis described in Section 2.5 (Fig. 5a,
 423 b). Overall, aerosol substantially influenced China’s terrestrial carbon uptake by altering radi-
 424 ation composition and meteorological conditions. At the national scale, the increase in diffuse
 425 radiation emerged as the dominant positive driver, contributing to GPP ($325.07 \text{ TgC yr}^{-1}$) and
 426 NEP ($11.46 \text{ TgC yr}^{-1}$). This highlights the crucial role of the diffuse radiation fertilization effect,
 427 particularly in regions with high aerosol loading across eastern and southwestern China, where
 428 enhanced diffuse light improves canopy light distribution and photosynthetic efficiency. In

429 contrast, the reduction in direct radiation suppressed GPP ($94.78 \text{ TgC yr}^{-1}$) and NEP (2.59 TgC
430 yr^{-1}) due to insufficient illumination, though the impact on NEP was weaker, reflecting partial
431 offset by the reduction in ecosystem respiration under aerosol-induced cooling. Cooling alone
432 reduced GPP by $59.62 \text{ TgC yr}^{-1}$ and NEP by 4.73 TgC yr^{-1} . This decline in NEP occurred
433 because the decrease in GPP (driven by reduced transpiration and stomatal conductance) out-
434 weighed the concurrent reduction in ecosystem respiration. Meanwhile, lower VPD enhanced
435 GPP by $114.44 \text{ TgC yr}^{-1}$ and NEP by 8.25 TgC yr^{-1} by alleviating water stress, reinforcing
436 photosynthetic carbon uptake. Changes in precipitation played only a minor role, slightly re-
437 ducing GPP (8.17 TgC yr^{-1}) and NEP (0.62 TgC yr^{-1}), with limited influence even in the mon-
438 soon regions of southern China. These findings indicate that variations in radiation components,
439 rather than hydrometeorological perturbations, serve as the primary pathway through which
440 aerosols modulate terrestrial carbon sinks.

441 Regionally, among these factors, diffuse radiation exerted the strongest positive influence
442 on GPP across all regions, particularly in the Southwest ($115.92 \text{ TgC yr}^{-1}$), East ($67.04 \text{ TgC yr}^{-1}$),
443 and South Central ($93.08 \text{ TgC yr}^{-1}$) China (Fig. 5a, b). Enhanced diffuse light under elevated
444 aerosol loading improved the vertical distribution of photosynthetically active radiation within
445 the canopy and increased photosynthetic efficiency. In contrast, direct radiation consistently
446 exhibited negative effects, most evident in the southwest ($-42.32 \text{ TgC yr}^{-1}$) and east (-22.92
447 TgC yr^{-1}), indicating that aerosol-induced solar dimming partly offset the diffuse radiation fer-
448 tilization benefit. Temperature changes associated with aerosol cooling suppressed GPP na-
449 tionwide, especially in the Southwest ($-29.07 \text{ TgC yr}^{-1}$) and South Central ($-16.19 \text{ TgC yr}^{-1}$),
450 by lowering canopy temperature and reducing evapotranspiration. The contributions of precip-
451 itation were minor (-1 to -3 TgC yr^{-1}), while VPD exerted a positive effect, particularly in
452 humid Southwest ($55.76 \text{ TgC yr}^{-1}$), suggesting that aerosol-induced cooling and moistening
453 alleviated water stress and indirectly promoted carbon uptake. For NEP, diffuse radiation re-
454 mained the dominant positive driver, with the largest increases in the Southwest (4.06 TgC yr^{-1})
455 and south-central (3.35 TgC yr^{-1}) China, while direct radiation continued to exert negative
456 effects. The temperature effect was moderate but consistent with GPP, implying that aerosol

457 cooling simultaneously suppressed photosynthesis and respiration, with a net positive outcome
 458 for NEP. Taken together, these spatial contrasts highlight the combined effects of aerosol com-
 459 position, vegetation structure, and regional hydroclimate, emphasizing that radiative forcing
 460 dominates in humid, high-biomass ecosystems, whereas climatic constraints prevail in arid
 461 zones.

462 To further clarify the dominant controls of these spatial differences, we identified the pri-
 463 mary meteorological drivers of GPP and NEP based on the standardized regression coefficients
 464 (Fig. 5c, d, and Table 2). The results indicate that for GPP, diffuse radiation accounts for the
 465 largest proportion (77.83%), followed by vapor pressure deficit (9.27%) and direct radiation
 466 (8.4%), while the influence of temperature (4.45%) and precipitation (0.05%) is relatively
 467 small. For NEP, diffuse radiation remains the dominant driver (72.20%), followed by direct
 468 radiation (15.92%) and temperature (5.64%). These results highlight that aerosols modify the
 469 radiation composition, particularly by enhancing diffuse radiation, which substantially in-
 470 creases photosynthetic efficiency and strengthens the regional carbon sink. In contrast, the ef-
 471 fects of temperature and VPD are weaker overall but more pronounced in northern arid and
 472 semi-arid regions, where water limitation constrains carbon uptake. Collectively, these findings
 473 confirm that radiation composition primarily controls the spatiotemporal dynamics of China’s
 474 terrestrial carbon sink, while temperature and moisture factors exert region-dependent modu-
 475 lations.

476

477 **Table 2.** Proportion of dominant meteorological factors for GPP and NEP across China
 478 (Units: %).

| Factors | RadD (%) | RadF (%) | Temp (%) | Precip (%) | VPD(%) |
|---------|----------|----------|----------|------------|--------|
| GPP | 8.40 | 77.83 | 4.45 | 0.05 | 9.27 |
| NEP | 15.92 | 72.20 | 5.64 | 0.14 | 6.10 |

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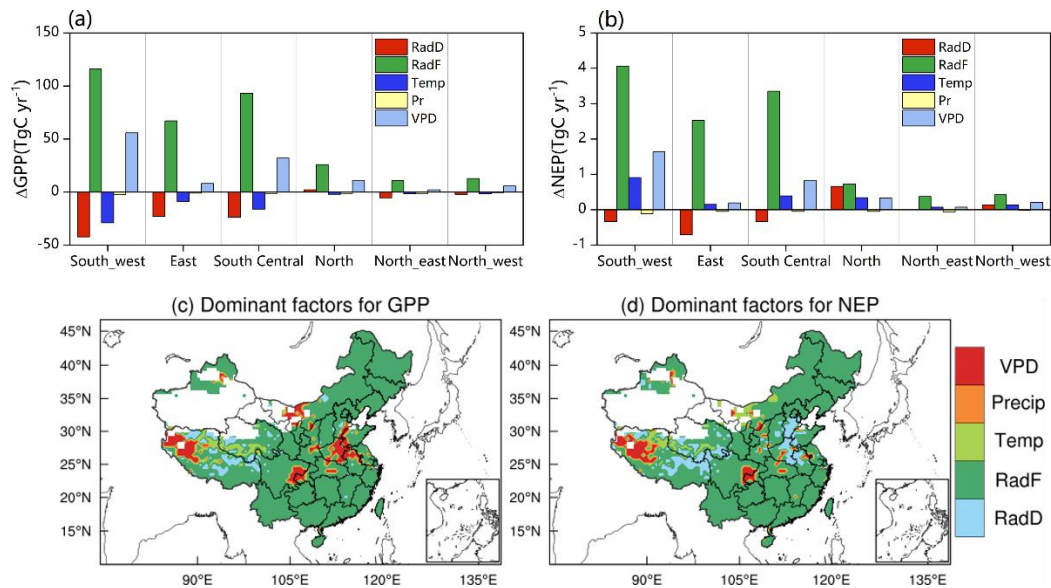
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487 **Figure 5.** Effect of aerosol-induced changes in meteorological factors on GPP and NEP, and
 488 spatial patterns of dominant factors. (a) Regional contributions of individual meteorological
 489 factors to GPP; (b) Regional contributions of individual meteorological factors to NEP; (c)
 490 Spatial distribution of the dominant meteorological factor for GPP; (d) Spatial distribution of
 491 the dominant meteorological factor for NEP.

492

493 3.3 Effects of surface ozone on carbon sinks

494 During 2010–2020, surface O₃ in China increased and imposed a persistent suppression
 495 on terrestrial carbon sinks. Simulations show a strong reduction of GPP by 0.4–0.6 gC m⁻² day⁻¹
 496 in most regions, with more than 0.8 gC m⁻² day⁻¹ in Southeast and Southwest China (Fig. 6a).
 497 NEP shows a similar spatial pattern (Fig. 6b). The largest decline occurs in the southeast (Yang-
 498 tze River basin and South China coast), with NEP reduced by 0.06–0.08 gC m⁻² day⁻¹ and
 499 locally above 0.1 gC m⁻² day⁻¹, consistent with high O₃ and evergreen broadleaf forests (Yue et
 500 al., 2017). In the southwest (Sichuan Basin and Yunnan–Guizhou Plateau), NEP decreases by
 501 0.03–0.06 gC m⁻² day⁻¹, related to complex terrain and dense forests. Impacts are weaker in
 502 Northeast and Northwest China, mostly below 0.02 gC m⁻² day⁻¹. In Shandong, Henan, and
 503 northern Jiangsu, the simulated losses are small, reflecting cropland-dominated land cover.

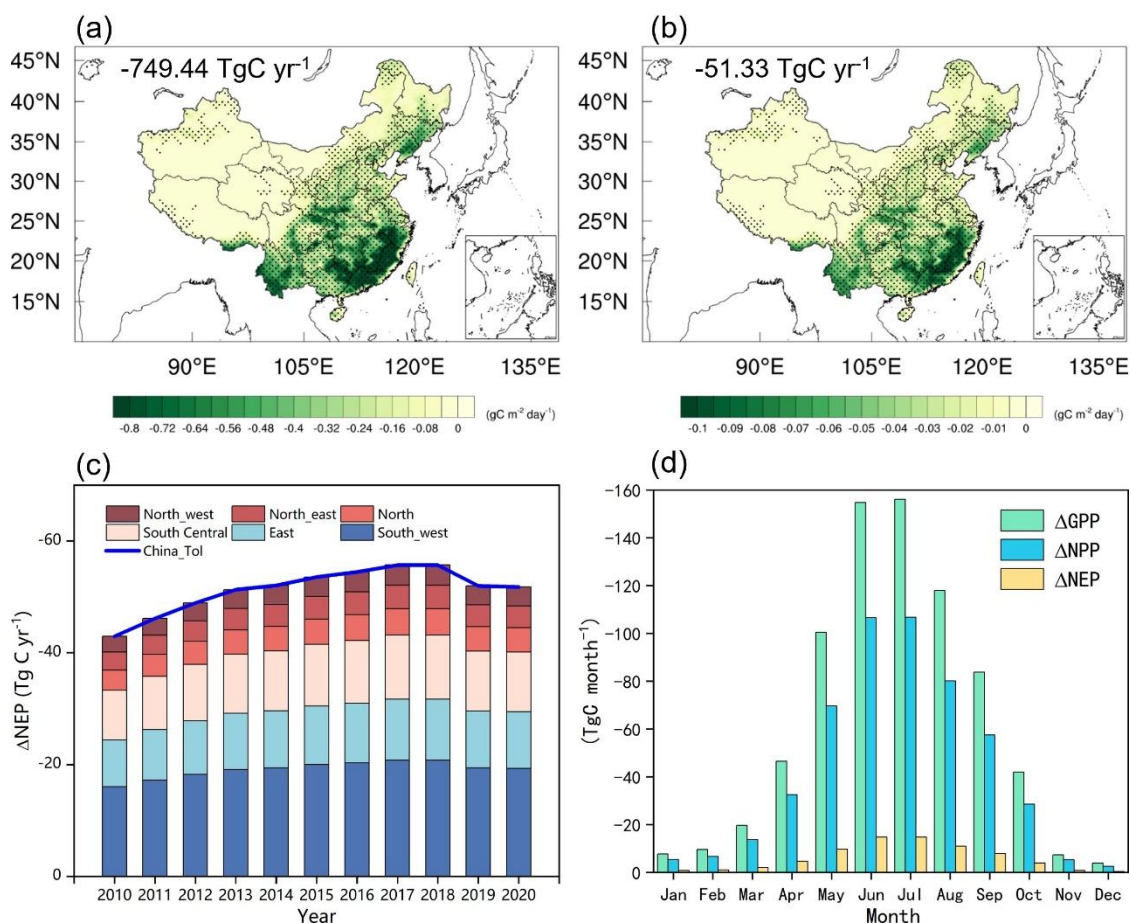
504 However, earlier studies reported strong O₃ effects on crops (Ren et al., 2012), suggesting possible underestimation. This bias may stem from the simplified crop representation in the model
505 (Fig. S2). Nationwide, O₃ reduces GPP and NEP by 749.44 TgC yr⁻¹ and 51.33 TgC yr⁻¹, accounting for 10.17 % and 12.90 % of the totals. O₃ also decreased Rh by 288.17 TgC yr⁻¹, with
506 the strongest reductions occurring in eastern and southern China (Fig. S10b). This indicates
507 that the O₃-induced suppression of ecosystem carbon uptake was partly offset by a concurrent
508 decline in heterotrophic respiration. This pattern suggests that reduced photosynthesis and carbon
509 allocation under O₃ stress decreased litter input and belowground carbon supply, thereby
510 limiting microbial substrate availability and weakening soil carbon decomposition. The suppression
511 is attributed to reduced photosynthesis, altered stomatal conductance, and shifts in
512 carbon allocation, which together weaken ecosystem sinks.

515 The annual effect intensifies until 2018 and then weakens (Fig. 6c). In 2010, O₃ reduces
516 NEP by 42.93 TgC yr⁻¹, reaching 55.71 TgC yr⁻¹ in 2018. It then decreases to 51.98 TgC yr⁻¹
517 in 2019 and 51.77 TgC yr⁻¹ in 2020. These variations reflect air pollution control policies. Between
518 2013 and 2017, the first Clean Air Action reduced PM_{2.5} and NO_x but left volatile organic
519 compounds (VOCs) largely uncontrolled, thereby enhancing O₃ formation, especially in
520 VOCs-limited regions (Lu et al., 2020). Both model and observations show higher O₃ during
521 this stage (Fig. S4). After 2018, the second Clean Air Action introduced coordinated control of
522 NO_x and VOCs in the Yangtze River Delta and Pearl River Delta, reducing O₃ during summer
523 and easing sink suppression in 2019–2020. In contrast, O₃ continued to rise in North China,
524 indicating uneven policy outcomes across regions.

525 Seasonal effects are distinct (Fig. 6d and Fig. S9). Summer shows the strongest suppression,
526 with NEP reduced by 29.10 TgC (56.69 % of the annual effect). This results from the
527 overlap of peak O₃ and peak photosynthesis, when high temperature and humidity keep stomata
528 open and allow O₃ uptake. Spring is second, with NEP reduced by 11.67 TgC (22.74 %). The
529 effect is linked to leaf expansion, rapid growth, and frequent transport events. Autumn and
530 winter show weaker impacts due to lower photosynthesis, unfavorable O₃ chemistry, and reduced
531 stomatal conductance. Regional differences are evident: in the south, suppression extends
532 from spring to late autumn, while in the north it is confined to summer. This highlights

533 the role of climate and phenology in modulating the impact of O₃ on carbon sinks.

534



535

536 **Figure 6.** Spatiotemporal variations in O₃-induced changes in carbon fluxes during 2010–2020.

537 (a–b) Multi-year mean spatial patterns of O₃-induced changes in GPP and NEP. National totals
538 are shown in each panel. Black dots denote significant changes ($p < 0.01$). (c) Interannual
539 variation of O₃-induced NEP. (d) O₃-induced monthly variations in GPP, NPP, and NEP.

540

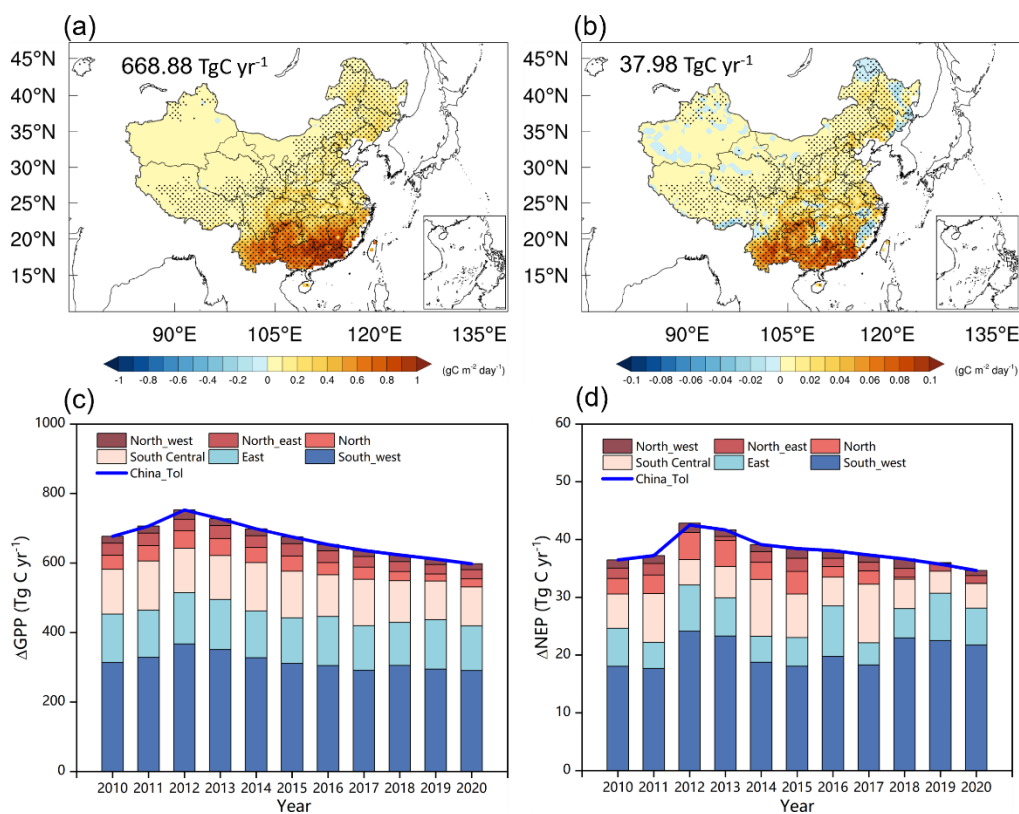
541 3.4 Effects of atmospheric nitrogen deposition on carbon sinks

542 The response of China's terrestrial ecosystems to atmospheric nitrogen deposition during
543 2010–2020 shows pronounced spatial heterogeneity (Fig. 7a, b). At the national scale, nitrogen
544 deposition increased GPP and NEP by 668.88 TgC yr⁻¹ and 37.98 TgC yr⁻¹, respectively. These
545 increases account for 9.08% of total GPP and 9.52% of total NEP. Atmospheric nitrogen dep-
546 osition also increased Rh by 297.26 TgC yr⁻¹ over China, indicating that the nitrogen-induced
547 enhancement of carbon uptake was accompanied by stronger soil carbon decomposition. The

548 net gains were mainly concentrated in the southeastern, southwestern, and central regions. In
549 these areas, NEP increased by $0.04\text{--}0.08\text{ g C m}^{-2}\text{ day}^{-1}$, forming the dominant contribution to
550 the nitrogen-induced carbon sink. Although atmospheric nitrogen deposition is highest in east-
551 ern China (Fig. 2e, f), the regional variations in GPP and NEP induced by nitrogen deposition
552 are more pronounced in southern China than in the east. The strong spatial gradient highlights
553 that the ecological effects of nitrogen deposition are not uniform, but tightly linked to anthro-
554 pogenic nitrogen emissions and ecosystem sensitivity (Shang et al., 2024). High responses were
555 observed in regions with intensive agriculture and industry, where deposition exceeded 15 kg
556 $\text{N ha}^{-1}\text{ yr}^{-1}$. Vegetation dominated by subtropical evergreen broadleaf forests, mixed forests,
557 and croplands is generally nitrogen-limited. Additional nitrogen input alleviated nutrient con-
558 straints, enhanced photosynthesis and biomass accumulation, and shifted soil microbial pro-
559 cesses. The spatial pattern of Rh (Fig. S10c) also shows pronounced positive responses in
560 southern China, consistent with the regions of strong nitrogen-induced carbon uptake. This
561 suggests that enhanced plant production and carbon input to soils stimulated microbial decom-
562 position, so that the final NEP gain reflects the balance between increased NPP and increased
563 Rh rather than a simple linear fertilization effect. When stimulation of GPP and NPP out-
564 weighed the increase in ER, NEP rose. Warm and humid climates, together with long growing
565 seasons, further amplified these effects.

566 The impacts of nitrogen deposition on GPP and NEP varied strongly over time (Fig. 7c,
567 d). In 2010, deposition enhanced NEP by 36.45 TgC yr^{-1} . The effect increased to a peak of
568 42.50 TgC yr^{-1} in 2012, but then declined, reaching 34.65 TgC yr^{-1} by 2020. This trajectory
569 reflects the influence of China's air pollution control policies on ecosystem carbon dynamics.
570 The temporal trend corresponds to changes in nitrogen deposition fluxes. Between 2010 and
571 2012, rapid industrialization and agriculture raised deposition from 15.85 to $17.91\text{ kg N ha}^{-1}$
572 yr^{-1} (+13%). After 2013, emission reduction policies reduced nitrogen deposition, which fell to
573 $13.25\text{ kg N ha}^{-1}\text{ yr}^{-1}$ in 2020 (-26.02%). Notably, the effect of nitrogen deposition on NEP
574 leveled off after 2015, which can be attributed to the slower decline rate of atmospheric nitro-
575 gen deposition since 2015 (Fig. S5). The reduction in NEP (-18.47%) was smaller than that in
576 nitrogen input. This lagged response suggests that soil nitrogen pools accumulated from long-

577 term deposition continued to supply nitrogen to vegetation, buffering the decline.

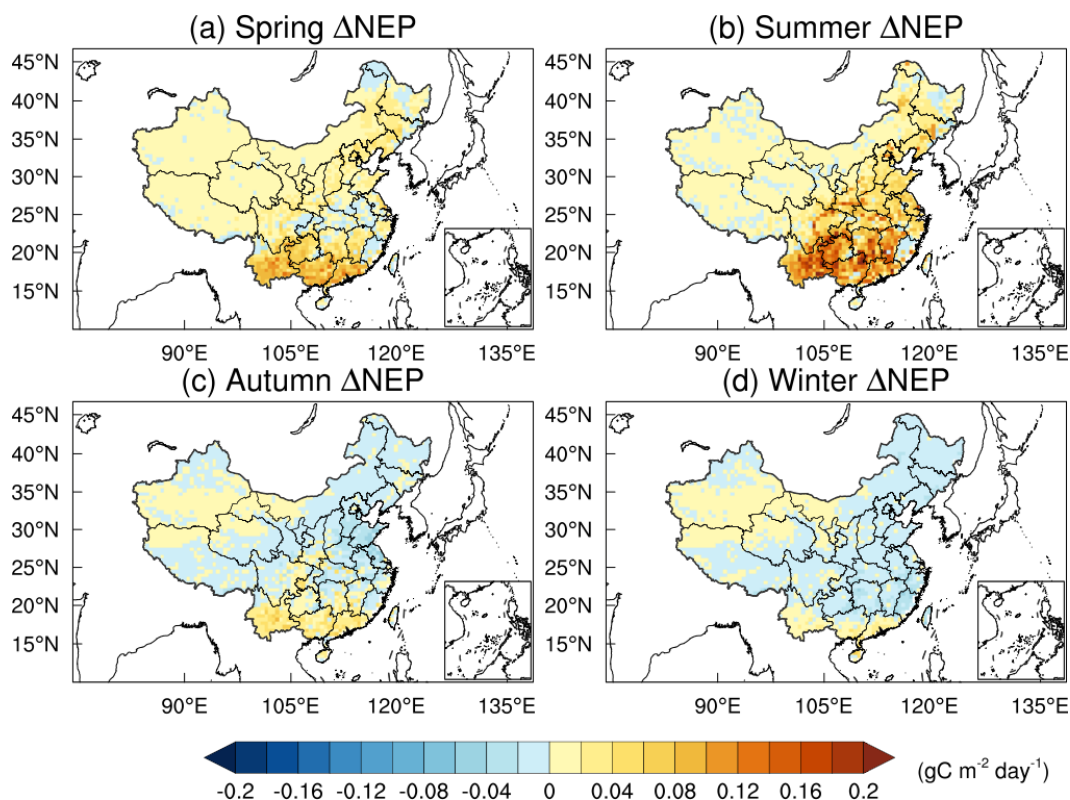


578
 579 **Figure 7.** Spatiotemporal variations in Ndep-induced changes in carbon fluxes during 2010–
 580 2020. (a–b) Multi-year mean spatial patterns of Ndep-induced changes in GPP and NEP. Na-
 581 tional totals are shown in each panel. Black dots denote significant changes ($p < 0.01$). (c-d)
 582 Interannual variation of Ndep-induced in GPP and NEP.

583
 584 The influence of nitrogen deposition on NEP displayed clear seasonality (Fig. 8). Strong
 585 positive effects occurred in summer and spring, whereas autumn and winter showed overall
 586 suppression at the national scale. Summer accounted for the largest gain, with an NEP increase
 587 of 27.16 TgC. Spring followed with 14.12 TgC. In contrast, autumn and winter reduced NEP
 588 by 0.20 and 3.10 TgC, respectively. These seasonal differences result from the combined influ-
 589 ence of multiple factors. During summer, optimal temperature, light, and water supported vig-
 590 orous canopy photosynthesis. Plants assimilated nitrogen efficiently, leading to higher GPP and
 591 biomass accumulation. Spring growth stages were also nitrogen-sensitive, producing strong
 592 positive responses. In autumn and winter, however, plant activity slowed. Nitrogen inputs
 593 mainly stimulated heterotrophic respiration, while GPP and NPP remained low. As a result,

594 NEP decreased, and carbon sink strength weakened outside the growing season. However, the
 595 negative response in autumn and winter was not spatially uniform. Positive NEP anomalies
 596 were still evident in parts of the eastern Qinghai–Tibet Plateau and western Sichuan, especially
 597 in autumn (Fig. 8c), whereas in winter this signal became much weaker and more localized
 598 (Fig. 8d). This spatial heterogeneity likely reflects the combined effects of persistent nitrogen
 599 limitation and weak heterotrophic respiration responses under cold, high-elevation conditions.
 600 In these regions, low temperatures constrain soil decomposition more strongly, so the stimula-
 601 tion of heterotrophic respiration by additional nitrogen is limited. Meanwhile, alpine grasslands,
 602 shrublands, and montane forests may still maintain some residual photosynthetic activity dur-
 603 ing the late growing season, allowing deposited nitrogen to support carbon uptake. As a result,
 604 nitrogen deposition can still enhance NEP locally in autumn, even though most other regions
 605 show seasonal carbon sink weakening.

606



607

608 **Figure 8.** Spatial distribution of Ndep-induced seasonal variations in NEP during 2010–2020
 609 (units: $\text{gC m}^{-2} \text{day}^{-1}$). (a) Spring, including March, April, and May. (b) Summer, including June,
 610 July, and August. (c) Autumn, including September, October, and November. (d) Winter, in-
 611 cluding January, February, and December.

612

613 **3.5 Integrated Impact Analysis**

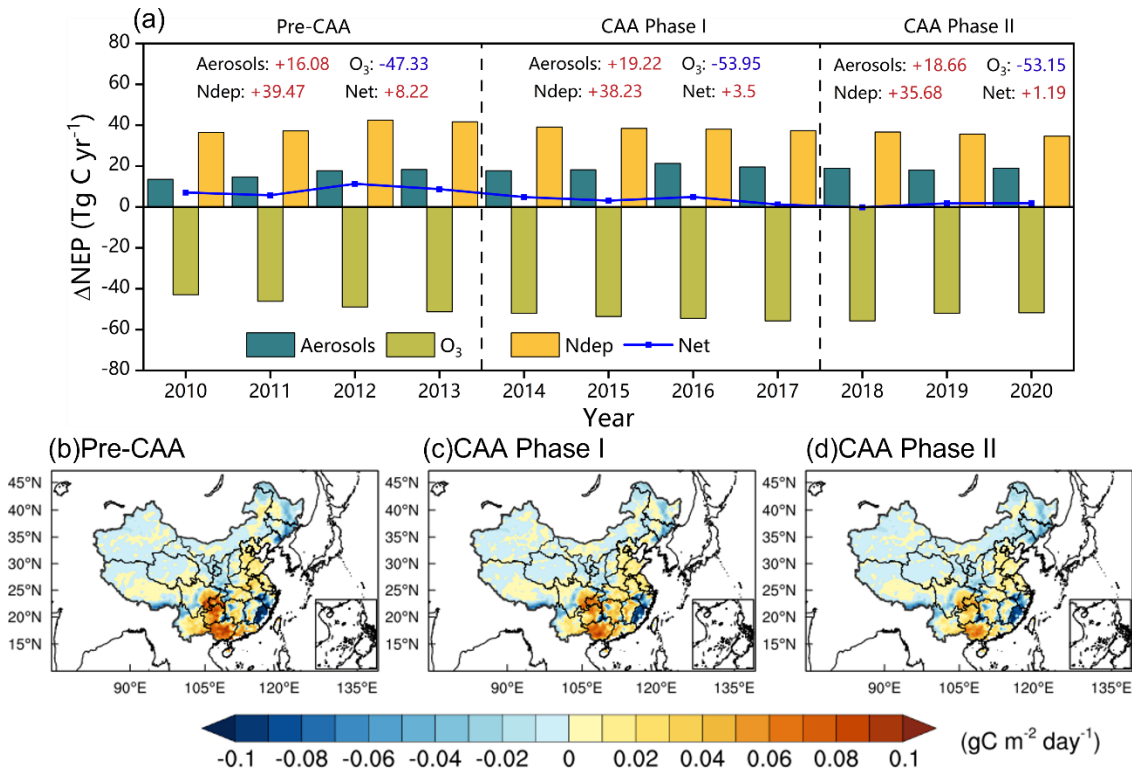
614 To assess the combined influence of aerosols, O₃, and atmospheric nitrogen deposition on
615 China's terrestrial carbon sink, the three independent effects were algebraically summed. Dur-
616 ing 2010–2020, the co-evolution of these atmospheric factors jointly drove substantial interan-
617 nual variability and stage-dependent changes in carbon uptake, closely linked to the implemen-
618 tation of the CAA plan. The interannual trend (Fig. 9) shows that although aerosols and nitrogen
619 deposition generally enhanced carbon sequestration, the strong carbon loss caused by O₃
620 largely offset these positive effects. The mean net effect was 4.58 TgC yr⁻¹, exhibiting pro-
621 nounced fluctuations and a declining trend. Net enhancement was strong in the early years of
622 the decade but weakened steadily and approached neutral levels by 2018–2020, when a slight
623 negative value (-0.14 TgC yr⁻¹) first appeared. These changes indicate a gradual transition from
624 an enhancement-dominated to an inhibition-dominated regime.

625 To further interpret this transition, the study period was divided into three phases accord-
626 ing to key CAA milestones, and the dominant factors were identified (Fig. 9a). In pre-CAA
627 (2010–2013), the mean annual net effect reached 8.22 TgC yr⁻¹, characterized by nitrogen-
628 deposition-dominated enhancement. Nitrogen deposition provided the largest positive contri-
629 bution (+39.47 TgC yr⁻¹), while the diffuse-radiation fertilization effect of aerosols offered a
630 secondary gain (+16.08 TgC yr⁻¹). The negative impact of O₃ (-47.33 TgC yr⁻¹) was largely
631 compensated by the two positive drivers, resulting in a pronounced increase in carbon sink
632 strength. This stage corresponded to relatively high emissions of aerosol and nitrogen precur-
633 sors, which maintained elevated aerosol loading and nitrogen deposition, while O₃ pollution
634 had not yet reached the stronger suppressive level observed in later years. During CAA Phase
635 I (2014–2017), the mean net effect decreased sharply to 3.50 TgC yr⁻¹, marking a transitional
636 stage with competing influences. The positive effect of aerosols peaked (+19.22 TgC yr⁻¹),
637 likely because aerosol changes during this period were more favorable for diffuse-radiation
638 fertilization, despite the concurrent declines in both scattering and absorbing aerosols. However,
639 this gain was largely offset by intensified O₃-induced inhibition (-53.80 TgC yr⁻¹). This phase

640 coincided with strong reductions in emissions of SO₂, PM_{2.5} and NO_x under the first Clean Air
641 Action (Fig. S11), which substantially altered atmospheric composition. Although declining
642 aerosol loading weakened the total radiative perturbation, the remaining aerosol conditions still
643 supported a strong diffuse-radiation effect. Meanwhile, insufficient VOCs control favored O₃
644 formation in many regions, thereby amplifying O₃-induced suppression of the carbon sink. In
645 CAA Phase II (2018–2020), the mean net effect further declined to 1.19 TgC yr⁻¹, forming an
646 O₃-dominated pattern. This stage was associated with further declines in aerosol concentrations
647 and nitrogen deposition under continued emission reductions, which weakened their positive
648 contributions to NEP. At the same time, the coordinated control of NO_x and VOCs in key re-
649 gions partly alleviated O₃ pollution, but this improvement was not sufficient to reverse the
650 dominant suppressive role of O₃ at the national scale. Overall, the stage-dependent changes in
651 the net carbon sink effect were broadly consistent with the temporal evolution of anthropogenic
652 emissions during 2010–2020 (Fig. S11), highlighting the strong imprint of CAA-related emis-
653 sion controls on the balance among aerosols, O₃, and nitrogen deposition. With continued emis-
654 sion control, the aerosol-induced enhancement decreased from its peak (+18.66 TgC yr⁻¹), and
655 the nitrogen-deposition gain weakened (+35.68 TgC yr⁻¹). Although O₃ suppression slightly
656 eased (-53.15 TgC yr⁻¹), it still nearly balanced the combined positive contributions, indicating
657 a fundamental shift in atmospheric drivers controlling China’s terrestrial carbon sink.

658 The spatial overlay further supports these findings (Fig. 9b, c, d). In forested and indus-
659 trialized regions of eastern and southern China, the cancellation between positive and negative
660 effects was most pronounced. These areas, benefiting from nitrogen and aerosol fertilization
661 but suffering from intense O₃ pollution, became hotspots of weakened or even reversed net
662 effects. Overall, the CAA plan not only improved air quality but also altered atmospheric com-
663 position in ways that substantially affected China’s terrestrial carbon sinks. Policy-driven emis-
664 sion changes transformed the system from a nitrogen–aerosol-enhanced regime to an O₃-dom-
665 inated offset pattern. These results suggest that achieving synergistic benefits between air-qual-
666 ity improvement and carbon neutrality requires elevating O₃ mitigation to a higher strategic
667 priority.

668



669

670 **Figure 9.** The overall impacts of aerosols, O₃, and atmospheric nitrogen deposition on the terrestrial carbon sink in China during 2010–2020. (a) Interannual variations of the combined
 671 effects. Pre-CAA represents the period before the implementation of the Clean Air Action
 672 (2010–2013); CAA Phase I and CAA Phase II represent the first (2014–2017) and second
 673 (2018–2020) stages of the CAA, respectively. (b–d) Spatial distributions of the annual means
 674 during the Pre-CAA, CAA Phase I, and CAA Phase II periods.
 675

676

677 3.6 Uncertainties

678 Although the RegESM framework captures the overall spatiotemporal variations of
 679 China’s terrestrial carbon sink in response to atmospheric composition changes, several uncer-
 680 tainties remain that may influence the quantitative assessment of the individual and combined
 681 effects of aerosols, O₃, and nitrogen deposition.

682 First, this study only considered the direct radiative effects of aerosols, while aerosol–
 683 cloud interactions were excluded. The first and second indirect effects of aerosols on cloud
 684 formation and albedo involve large uncertainties (Haywood and Boucher, 2000) and were
 685 therefore not represented in our simulations. However, observations have shown that terrestrial

686 carbon fluxes are highly sensitive to sky conditions and diffuse radiation changes (Oliphant et
687 al., 2011; Yue and Unger, 2017). The omission of aerosol–cloud interactions may affect the
688 magnitude and spatial pattern of aerosol impacts on radiation and photosynthesis, as cloud-
689 mediated diffuse radiation responses remain uncertain. Future work should explicitly include
690 aerosol–cloud–radiation feedbacks to better quantify their effects on ecosystem carbon ex-
691 change.

692 Second, uncertainties remain in evaluating vegetation responses to O₃ exposure. Field-
693 based O₃ fumigation experiments across China are still limited, making it difficult to compre-
694 hensively assess ecosystem-level damage. In this study, the YIBs model applied different O₃
695 damage coefficients for plant functional types, and the parameterization has shown reasonable
696 regional performance in simulating GPP–O₃ responses (Yue et al., 2017). Nevertheless, a wider
697 range of site-level observations is required to constrain the O₃ damage functions across various
698 vegetation types and climate zones at the national scale.

699 Third, nonlinear coupling among aerosols, O₃, and nitrogen deposition introduces sys-
700 temic uncertainty in estimating their combined effects. Aerosol reduction alters photolysis rates
701 and thereby affects O₃ formation (Tang et al., 2017; Yan et al., 2023; Yang et al., 2022), while
702 O₃ and nitrogen jointly regulate stomatal conductance, photosynthetic efficiency, and water-
703 use dynamics (Sitch et al., 2007; Zhang et al., 2018). In addition, the coexisting effects of these
704 three drivers may not be strictly additive. Aerosol effects and O₃ damage may be partly antag-
705 onistic, as aerosol-induced changes in diffuse radiation, temperature, and vapor pressure deficit
706 can modulate stomatal uptake and the physiological stress caused by O₃. Nitrogen deposition
707 may partly offset O₃-induced reductions in plant productivity by alleviating nutrient limitation,
708 whereas the interaction between aerosols and nitrogen deposition may vary regionally because
709 nitrogen availability can influence the extent to which vegetation benefits from aerosol-induced
710 radiation changes. These interactions may amplify or offset each other under changing climatic
711 conditions, and the real-world coexisting effects of these atmospheric drivers may therefore
712 differ from the linear sum of their independently quantified contributions. In the present study,
713 the three factors were quantified separately to isolate their first-order effects, while a full as-
714 sessment of their two-way and three-way interactions would require a dedicated factorial

715 experimental framework. Therefore, although nonlinear interactions may modify the quantita-
716 tive magnitudes of the simulated responses, the overall conclusion that the positive contribu-
717 tions of aerosols and nitrogen deposition weakened while the suppressive influence of O₃ be-
718 came increasingly important during 2010–2020 is expected to remain qualitatively robust.
719 These limitations further emphasize the need for high-resolution, fully coupled chemistry–eco-
720 system modeling frameworks to capture the co-evolution of multiple atmospheric processes.

721 In summary, despite these uncertainties, this study provides robust quantitative evidence
722 that aerosols, O₃, and nitrogen deposition jointly modified the magnitude and spatial distribu-
723 tion of China’s terrestrial carbon sink during 2010–2020. Future efforts should focus on incor-
724 porating aerosol–cloud interactions, expanding field-based O₃ response networks, and improv-
725 ing representation of multi-process coupling and nonlinear interactions to further constrain at-
726 mospheric–biosphere feedbacks under China’s evolving air quality and carbon neutrality goals.

727

728 **4 Conclusions**

729 This study employed the RegESM to quantify the effects of aerosol, surface O₃, and ni-
730 trogen deposition on China’s terrestrial carbon sink during 2010–2020. The model effectively
731 reproduced the spatial and temporal variations of aerosol optical depth, O₃, nitrogen deposition,
732 and carbon fluxes, providing a solid basis for process-level attribution analysis.

733 Aerosols exerted a substantial positive influence on China’s terrestrial carbon sink. On
734 average, aerosols enhanced GPP and NEP by 293.28 TgC yr⁻¹ (3.98%) and 17.93 TgC yr⁻¹
735 (4.49%), respectively, primarily through the diffuse radiation fertilization effect. The strongest
736 enhancement appeared in southern and eastern China, where high aerosol loading and dense
737 vegetation synergistically improved canopy light-use efficiency. Aerosol-induced surface cool-
738 ing and reduced VPD further alleviated water stress and stimulated carbon uptake. The en-
739 hancement peaked during 2015–2017, coinciding with elevated diffuse radiation fractions, and
740 weakened slightly under cleaner atmospheric conditions after 2018.

741 In contrast, surface O₃ persistently suppressed ecosystem carbon uptake, reducing GPP
742 and NEP by 749.44 TgC yr⁻¹ (10.17%) and 51.33 TgC yr⁻¹ (12.90%), respectively. The

743 strongest suppression occurred in southeastern and southwestern China, where dense forest
744 ecosystems coincided with high O₃ concentrations. O₃-induced damage peaked in 2018, con-
745 sistent with the exceptionally high O₃ levels. Subsequent coordinated NO_x–VOCs manage-
746 ment under the second Clean Air Action partially mitigated O₃ levels and NEP suppression. O₃
747 exerted a strongly seasonal negative impact on NEP, with the strongest suppression occurring
748 in summer.

749 Atmospheric nitrogen deposition enhanced the terrestrial carbon sink by 9.08% for GPP
750 and 9.52% for NEP, with effects concentrated in southern and central China. The enhancement
751 peaked around 2012, declined gradually after 2013 following reduced anthropogenic emissions,
752 and leveled off after 2015, corresponding to a slower decline in deposition and a lagged eco-
753 system response due to soil nitrogen accumulation. Seasonal variations showed stronger stim-
754 ulation in summer and spring, while autumn and winter exhibited minor reductions linked to
755 enhanced respiration.

756 During 2010–2020, the combined effects of aerosols, surface O₃, and atmospheric nitro-
757 gen deposition on China’s terrestrial carbon sink exhibited marked interannual variability and
758 a distinct transition under the Clean Air Action (CAA). The net atmospheric contribution de-
759 clined from +8.22 Tg C yr⁻¹ during the Pre-CAA period (2010–2013) to +1.19 Tg C yr⁻¹ in
760 Phase II (2018–2020), as the increasing suppression from O₃ (-53.15 Tg C yr⁻¹) gradually offset
761 the positive impacts of aerosols and nitrogen deposition. These results indicate that China’s air-
762 pollution control not only improved air quality but also altered atmospheric chemical compo-
763 sition in ways that significantly affected ecosystem carbon uptake, with O₃ becoming the dom-
764 inant limiting factor in the later period.

765 Overall, aerosols, O₃, and nitrogen deposition exerted interconnected yet contrasting in-
766 fluences on China’s terrestrial carbon sink. Aerosols and nitrogen deposition enhanced carbon
767 uptake through diffuse radiation and nutrient input, whereas O₃ caused physiological damage
768 that suppressed it. The evolving interplay among these factors illustrates how emission reduc-
769 tions, atmospheric chemistry, and ecosystem feedbacks jointly impact carbon sink dynamics
770 under China’s clean-air policies. These findings suggest that future air-quality management
771 should move beyond single-pollutant control and place greater emphasis on coordinated multi-

772 pollutant strategies, particularly the synergistic reduction of NO_x and VOCs, to limit O₃-in-
773 duced carbon sink suppression while sustaining gains in air quality. In addition, ecosystem
774 carbon-sink responses should be incorporated into the evaluation of clean-air policies, espe-
775 cially in ecologically sensitive regions of eastern and southern China, to better achieve co-
776 benefits for air pollution mitigation and carbon neutrality goals.

777 **Data Availability Statement**

778 MODIS data are provided at <https://doi.org/10.5067/MODIS/MOD17A2H.006> (last access: 11
779 May 2024; Running et al., 2015). AOD data come from the MODIS Level-3 monthly product
780 (MOD08_M3, Collection 6.1) available at https://doi.org/10.5067/MODIS/MOD08_M3.061
781 (last access: 5 November 2024; Platnick et al., 2015). WDCGG data are available at
782 <https://doi.org/10.15138/wkgj-f215> (last access: 10 June 2024; Lan, 2023). ERA-Interim data
783 are available at <https://doi.org/10.24381/cds.f2f5241d> (last access: 8 June 2024; Dee et al.,
784 2011). The Carbon Tracker data can be accessed at <https://gml.noaa.gov/ccgg/carbontracker/>
785 (last access: 21 June 2024; Jacobson et al., 2023). CNEMC data are provided at
786 <https://www.cnemc.cn/en> (Kong et al., 2021).

787 **Author contributions**

788 TW and NX designed the study. NX performed the analysis, conducted the evaluation, and
789 drafted the manuscript. TW supervised the research and acquired funding. Both authors con-
790 tributed to the interpretation of results and to reviewing and editing the manuscript.

791 **Competing interests**

792 The corresponding author has stated that all the authors have no conflicts of interest.

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