Elevated CO$_2$ negates O$_3$ impacts on terrestrial carbon and nitrogen cycles

Graphical abstract

Highlights

- e[O$_3$] decreases terrestrial C and N pools and fluxes

- e[CO$_2$] increases terrestrial C and N pools and fluxes

- e[CO$_2$] negates e[O$_3$] impacts on terrestrial C and N pools and fluxes

- Evaluating terrestrial C and N feedbacks to concurrent climate changes is critical
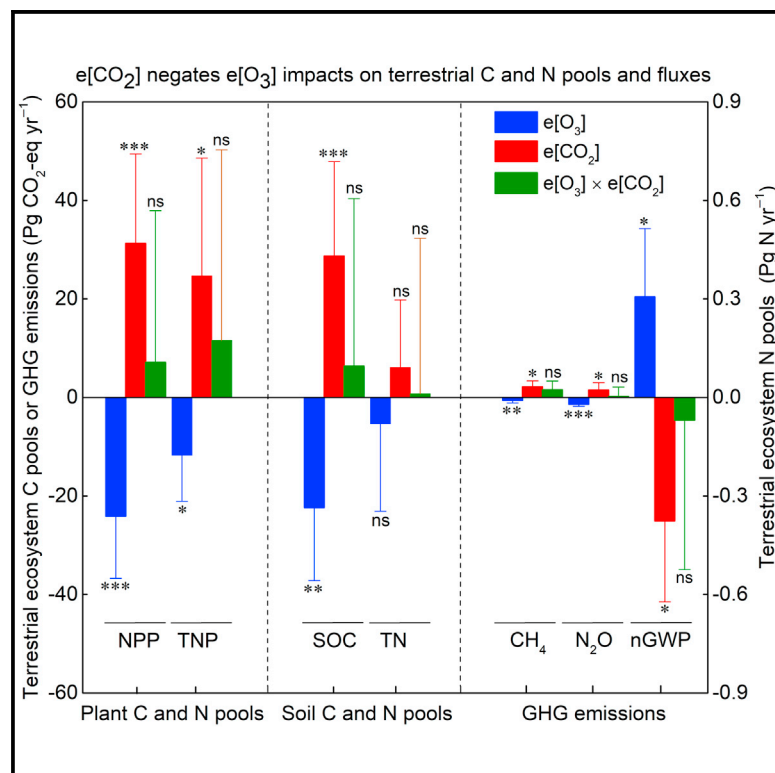
Authors

Longlong Xia, Shu Kee Lam,
Ralf Kiese, ..., Lei Ma, Yuhao Zhu,
Klaus Butterbach-Bahl

Correspondence

longlong.xia2@kit.edu (L.X.),
klaus.butterbach-bahl@kit.edu (K.B.-B.)

In brief

Increases in tropospheric concentrations of O$_3$ and CO$_2$ substantially alter the functions of global terrestrial ecosystems and elements cycles. Here, we synthesize global datasets and find that increased O$_3$ concentration significantly decreases terrestrial net primary productivity and various carbon and nitrogen pools and fluxes, which are greatly stimulated by increased CO$_2$ concentration. Moreover, increases in CO$_2$ concentration negate or even overcompensate the detrimental effects of increased O$_3$ concentration on terrestrial ecosystem functions and carbon and nitrogen cycles.

CellPress

Elevated CO$_2$ negates O$_3$ impacts on terrestrial carbon and nitrogen cycles

Longlong Xia,[1,13,*] Shu Kee Lam,[2] Ralf Kiese,[1] Deli Chen,[2] Yiqi Luo,[3] Kees Jan van Groenigen,[4] Elizabeth A. Ainsworth,[5,6,7] Ji Chen,[8] Shuwei Liu,[9,10] Lei Ma,[11] Yuhao Zhu,[1] and Klaus Butterbach-Bahl[1,12,*]

[1]Institute for Meteorology and Climate Research (IMK-IFU), Karlsruhe Institute of Technology, Garmisch-Partenkirchen 82467, Germany
[2]School of Agriculture and Food, Faculty of Veterinary and Agricultural Sciences, The University of Melbourne, Melbourne, VIC 3010, Australia
[3]Center for Ecosystem Science and Society, Department of Biological Sciences, Northern Arizona University, Flagstaff, AZ 86011, USA
[4]Department of Geography, College of Life and Environmental Sciences, University of Exeter, Exeter EX4 4RJ, UK
[5]Global Change and Photosynthesis Research Unit, USDA ARS, Urbana, IL 61801, USA
[6]Department of Plant Biology, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA
[7]Institute for Genomic Biology, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA
[8]Department of Agroecology, Aarhus University, Tjele 8830, Denmark
[9]Jiangsu Key Laboratory of Low Carbon Agriculture and GHGs Mitigation, College of Resources and Environmental Sciences, Nanjing Agricultural University, Nanjing 210095, China
[10]Jiangsu Key Lab and Engineering Center for Solid Organic Waste Utilization, Jiangsu Collaborative Innovation Center for Solid Organic Waste Resource Utilization, Nanjing Agricultural University, Nanjing 210095, China
[11]College of Atmospheric Sciences, Lanzhou University, Lanzhou 730000, China
[12]International Livestock Research Institute, Old Naivasha Road, Nairobi 30709, Kenya
[13]Lead contact
*Correspondence: longlong.xia2@kit.edu (L.X.), klaus.butterbach-bahl@kit.edu (K.B.-B.)
https://doi.org/10.1016/j.oneear.2021.11.009

SCIENCE FOR SOCIETY Plants, through various ecosystem functions, provide society with vital services, including important carbon sinks to combat climate change and a crucial food source. Over the past 100 years, atmospheric concentrations of two greenhouse gases that impair (ozone [O$_3$]) and enhance (carbon dioxide [CO$_2$]) plant growth have strongly increased. However, little is known about their net effects on plant traits and the associated ecosystem functions and services. In this work, we synthesize current knowledge and show that increased CO$_2$ concentration negates or even overcompensates the detrimental effects of increased O$_3$ on ecosystem functions and carbon and nitrogen cycles. Our assessment reveals the complex interactive impacts of changes in two key atmospheric constituents on terrestrial ecosystems and highlights the need to understand the complex effects of atmospheric composition changes on ecosystems while assessing options to mitigate human perturbation of the global environment.

SUMMARY

Increasing tropospheric concentrations of ozone (e[O$_3$]) and carbon dioxide (e[CO$_2$]) profoundly perturb terrestrial ecosystem functions through carbon and nitrogen cycles, affecting beneficial services such as their capacity to combat climate change and provide food. However, the interactive effects of e[O$_3$] and e[CO$_2$] on these functions and services remain unclear. Here, we synthesize the results of 810 studies (9,109 observations), spanning boreal to tropical regions around the world, and show that e[O$_3$] significantly decreases global net primary productivity and food production as well as the capacity of ecosystems to store carbon and nitrogen, which are stimulated by e[CO$_2$]. More importantly, simultaneous increases in [CO$_2$] and [O$_3$] negate or even overcompensate the negative effects of e[O$_3$] on ecosystem functions and carbon and nitrogen cycles. Therefore, the negative effects of e[O$_3$] on terrestrial ecosystems would be overestimated if e[CO$_2$] impacts are not considered, stressing the need for evaluating terrestrial carbon and nitrogen feedbacks to concurrent changes in global atmospheric composition.

INTRODUCTION

Since the Industrial Revolution, anthropogenic activities have been changing global atmospheric composition (e.g., increasing ozone and carbon dioxide concentrations).[1,2] This has profoundly disturbed terrestrial ecosystem functions that provide services vital to human life and health, such as combating climate change through sustaining ecosystem carbon storage and safeguarding food security.[1] The increased emissions of nitrogen oxides (NO_x) and volatile organic compounds (VOCs) in conjunction with fossil fuel burning have increased the global tropospheric ozone (O_3) concentrations by 40% since preindustrial times.[2] As a strong oxidant and air pollutant, O_3 enters plant leaves through the stomata, generating reactive oxygen species and causing oxidative stress,[3,4] which in turn reduces leaf photosynthesis, plant biomass, and crop yield.[4–6] A previous study suggests that an elevated O_3 concentration (e[O_3]) significantly decreased tree photosynthesis by 12%, shoot biomass by 9%, and root biomass by 12%.[7] This decrease in plant productivity may slow down the carbon (C) and nitrogen (N) cycling in terrestrial ecosystems.[8–11]

Decreased aboveground (ANPP) and belowground net primary productivity (BNPP) under e[O_3] result in lower C emissions through ecosystem respiration,[12,13] and less soil C inputs through leaf and root litter, thereby likely decreasing soil organic C (SOC) and N stocks.[14,15] Elevated [O_3] can also directly impose physiological stress on soil microorganisms and may decrease soil C turnover (e.g., SOC decomposition) and N transformation rates (e.g., mineralization, nitrification, and denitrification),[8,16–18] which in turn down-regulates the size of soil labile C and N pools and emissions of greenhouse gases (GHGs) from soils (carbon dioxide [CO_2], methane [CH_4], and nitrous oxide [N_2O]).[19–21] Recent studies have examined the response of ecosystem C pools to e[O_3] in forest ecosystems,[15,21,22] but are largely limited to C components (e.g., ANPP and BNPP). So far, no study has comprehensively assessed the effects of e[O_3] on various C and N pools in global terrestrial ecosystems, which limits our understanding of the impact of future [O_3] on terrestrial C and N cycles.

In parallel to e[O_3], atmospheric CO_2 concentrations have increased by nearly 47% since preindustrial times.[2] In contrast with e[O_3], elevated CO_2 concentrations (e[CO_2]) stimulate plant photosynthesis, promote NPP[23,24] and accelerate terrestrial C and N cycling.[25–27] Recent meta-analyses indicate that e[CO_2] significantly increased plant and soil C pools by 4%–23% in terrestrial ecosystems.[28,29] These results raise the question of whether e[CO_2] could overcome the negative effects of e[O_3] on terrestrial C and N pools.[5,15,22] Only a few studies have examined the interactive effects of e[CO_2] × e[O_3], but only on plant C pools in forest ecosystems and with limited data,[22,30] whereas the effects on N pools and other ecosystem types such as cropland and grassland were not considered. For example, tree ANPP was reported to be significantly decreased by 11%–16% under [O_3], but was increased by 12%–21% under e[O_3] × e[CO_2],[22] indicating an overcompensating effect of e[CO_2] on plant C pools. Whether this net positive effect also exists for plant N pools, soil C and N pools, and the associated GHG emissions in terrestrial ecosystems remains unclear. Moreover, it remains unknown whether the interactive effects of e[O_3] and

e[CO_2] on plant and soil C and N pools are additive, antagonistic, or synergistic. This uncertainty makes it difficult to predict the response of terrestrial C and N cycles to multiple global change drivers.[27,31] Addressing these knowledge gaps will reveal the capacity of terrestrial ecosystems to sustain global feed and food production and combat global warming under increasing O_3 and CO_2 concentrations.[32,33] Moreover, it will guide the development of ecosystem-based climate change mitigation strategies to improve the resilience of terrestrial ecosystems to concurrent changes in atmospheric composition and reduce the risks of climate change impacts on global environment and humans (e.g., heat waves and flooding).[32,33]

Here, we synthesized the results of 810 studies with 9,109 observations (see experimental procedures and Figure S1) to (1) quantify the effects of e[O_3], e[CO_2], and e[O_3] × e[CO_2] on key C and N cycling variables (viz. plant photosynthesis, plant biomass, crop yield, NPP, plant N pools, soil organic and labile C and N pools, ecosystem and soil respiration and GHG emissions) in global terrestrial ecosystems; (2) evaluate the interactive effects of e[O_3] and e[CO_2] on these C and N variables; and (3) explore whether e[CO_2] can overcome the negative effects of e[O_3] on terrestrial C and N pools. We found that e[O_3] and e[CO_2] exerted opposite effects on the targeted C and N cycling variables, and the interactive effects of e[O_3] and e[CO_2] were mostly additive. Simultaneous increases in [CO_2] and [O_3] negated or even overcompensated the negative effects of e[O_3] on terrestrial C and N pools and fluxes. These results highlight the importance of evaluating terrestrial C and N feedbacks to concurrent climate changes.

RESULTS AND DISCUSSION

Elevated O_3 and CO_2 concentrations

In total, 810 peer-reviewed publications (9,109 observations) reporting results from global terrestrial ecosystems, spanning boreal to tropical regions, were included in the meta-analysis (Figure S1). Most studies included in our dataset were conducted in the northern hemisphere; only 32 studies were conducted in the southern hemisphere. Across all studies included in our dataset, ambient atmospheric O_3 and CO_2 concentrations ranged between 15 and 50 ppbv (on average 36 ppbv) and 340–410 ppmv (on average 388 ppmv), respectively, while those of elevated O_3 and CO_2 concentrations ranged between 29 and 150 ppbv (on average 72 ppbv) and 400–880 ppmv (on average 615 ppmv). Models predict that the atmospheric [O_3] and [CO_2] will increase to 70–80 ppbv and 560–720 ppmv (RCP4.5–6.0) by 2100,[1] respectively. Thus, the magnitudes of [O_3] and [CO_2] enrichment of the studies in our dataset are consistent with projections for the end of this century.

Effects of e[O_3] on plant NPP and N pools

Averaged across all studies in our dataset, e[O_3] significantly decreased leaf net photosynthetic rate (P_n) by 26.0% compared with ambient [O_3] (Figures 1D and S6). The strong oxidative stress due to e[O_3] may cause leaf cell damage and decrease the supply of carbohydrate precursors,[4,5] which caused significantly decrease in stomatal conductance (g_s), carboxylation efficiency (V_{cmax}), and leaf chlorophyll content (Chll) (Figure 1D). This explains the observed decreases in photosynthesis and
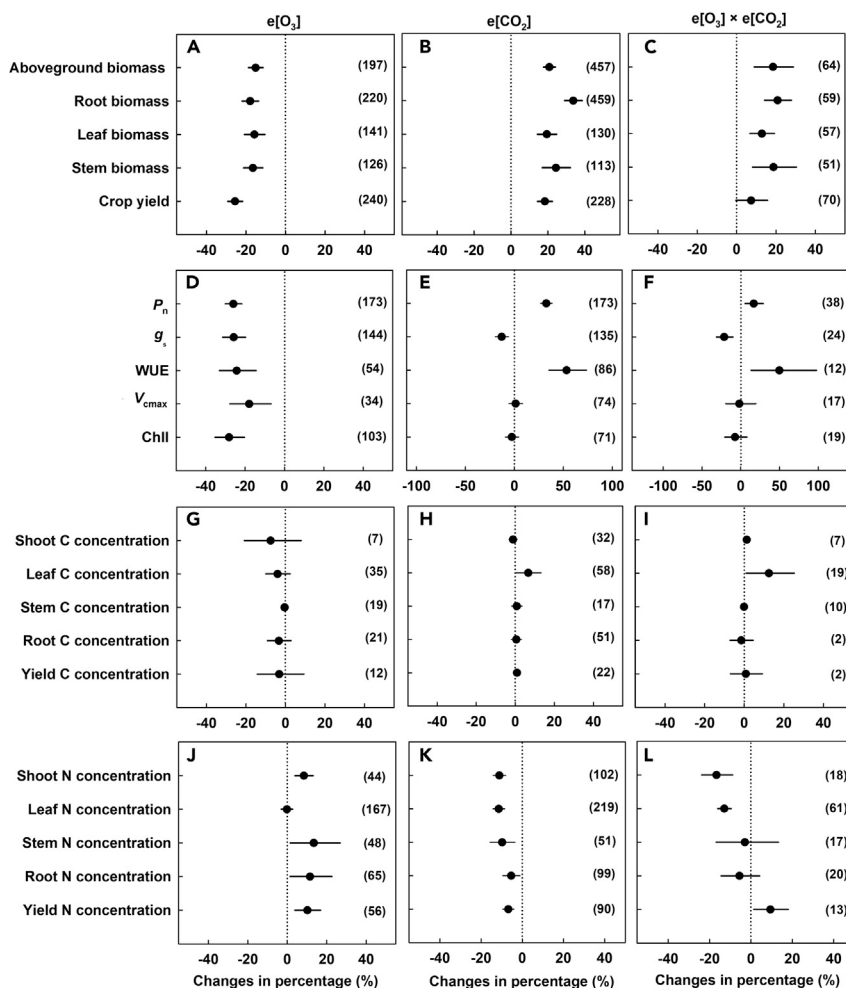
Figure 1. Responses of plant growth and photosynthesis to e[O₃], e[CO₂], and e[O₃] × e[CO₂]

Effects of elevated O₃ (e[O₃], on average 72 ppbv), elevated CO₂ (e[CO₂], on average 615 ppmv) and e[O₃] (on average 67 ppbv) × e[CO₂] (on average 612 ppmv) on plant biomass (A, B, and C), plant photosynthesis (D, E, and F), plant C (G, H, and I), and N concentrations (J, K, and L) in global terrestrial ecosystems. P_n, net photosynthetic rate; V_{cmax}, the maximum carboxylation rate; WUE, water use efficiency. Yield N concentration denotes the N concentration in the harvest part in croplands. The number of experimental observations is in parentheses.

accelerated leaf senescence. As a result, the biomass of plant tissues (shoot, root, leaf, and stem) was significantly decreased by 15.1%–17.9% (Figures 1A and S5; Table S6). Root growth was more negatively affected by e[O₃] than aboveground biomass as evidenced by the decreased root/shoot ratio (Figure S2). These results might be explained by the fact that leaves in the lower and upper canopies were the preferential sources of photosynthesis for root and shoot growth, respectively.[34] As e[O₃] accelerates the senescence of older (mature) leaves in the lower canopy, less C is allocated to roots, which explains the observed reductions of root/shoot ratio.[7] Compared with the varied responses of leaf P_n resulting from the vertical gradient of light intensity, the decrease in canopy photosynthesis (P_c) may better reflect the damage of e[O₃] on the capacity of plant assimilation of atmospheric CO₂.[3,35] P_c was not assessed in this study due to data deficiency. However, a previous study found that the P_c decreased significantly with increasing [O₃] and the decreasing rate (percentage reduction per unit [O₃] increase) was similar to the magnitude of decrease in plant aboveground biomass under e[O₃].[35]

Across all studies, e[O₃] significantly decreased crop yield by 25.6% (Figure 1A), which is within the range of reduction (23%–29%) reported in previous meta-analyses.[36,37] Moreover, the

decrease in crop yield increased significantly with the level of [O₃] enrichment (Figure 3). Model simulations suggest that e[O₃] decreased global crop yield by 227 Tg year⁻¹ (1 Tg = 10¹² g) during 2010–2012,[38] accounting for 9% of global crop production. These results highlight the urgency to implement measures to lower tropospheric O₃ concentrations, or at least to halt its increase, and to develop O₃-tolerant crop cultivars to safeguard global food security.[39]

Beside plant biomass and crop yield, plant NPP and its components (ANPP, BNPP, leaf, and stem C pools) were also significantly reduced by 21.6%–28.6% by e[O₃] (Figures 2A and S9) because of the decreased plant biomass and unaffected plant C concentrations (Figures 1A, 1G, and S7). Interestingly, e[O₃] significantly increased N concentrations in plant components (except for leaf) (Figures 1J and S8) and decreased plant C/N ratios (Figure S2). This might be explained by the decreased plant biomass relative to the amount of available N[5,40] or by N retranslocation from prematurely senescing leaves under e[O₃].[41] However, the increase in plant N concentrations was negligible compared with the magnitude of plant biomass reduction under e[O₃], which led to significant decreases in various plant N pools (10.0%–14.0%) (Figures 2D and S10).

We extrapolated our results on ecosystem C and N pools and fluxes to the global scale by multiplying averaged area-scaled effect sizes with the corresponding total land area (see experimental procedures). We found that e[O₃] significantly decreased plant NPP and total N pool (TNP) by 24.1 (95% confidence interval [CI], 11.5–36.7) Pg CO₂-equivalent (CO₂-eq) year⁻¹ and 0.18 (95% CI, 0.03–0.32) Pg N year⁻¹, respectively, in terrestrial ecosystems (Figures 2 and 6). The consistent decrease in plant NPP and N pools across all terrestrial ecosystems analyzed suggests that e[O₃] significantly reduces the capacity of plant biomass to store C and N. More importantly, the magnitude of decrease in plant NPP and N pools increased significantly with the level of [O₃] enrichment (Figure 3), suggesting that the losses in terrestrial plant C and N storage would be aggravated if [O₃] continues to increase.[5,42]
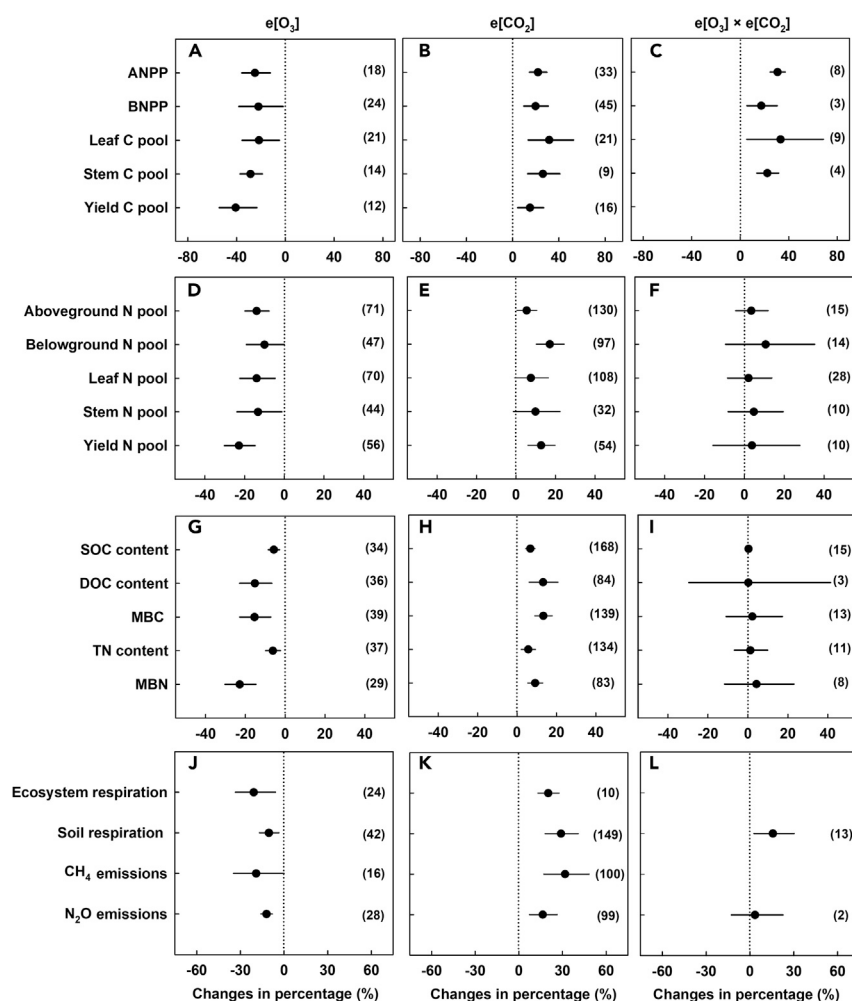
Figure 2. Responses of C and N pools and fluxes to e[O₃], e[CO₂], and e[O₃] × e[CO₂]

Effects of elevated O_3 (e[O_3], on average 72 ppbv), elevated CO_2 (e[CO_2], on average 615 ppmv) and e[O_3] (on average 67 ppbv) × e[CO_2] (on average 612 ppmv) on plant NPP components (A, B, and C) and N pools (D, E and F), and soil C and N pools (G, H, and I) and greenhouse gas emissions (J, K, and L) in global terrestrial ecosystems. CH_4 emissions represent the average emissions from paddy fields and wetlands. The number of experimental observations is in parentheses.

Effects of e[O₃] on soil C and N pools and GHG emissions

As the largest C reservoir in terrestrial ecosystems, soils (0–1 m layer) store about 1,500 Pg (1 Pg = 10^{15} g) of organic C,[46] which is 3–4 times more than that stored in atmospheric and plant pools.[47] SOC storage depends on the balance between soil C input (e.g., root and leaf litters) and output (soil respiration).[26,48] e[O_3] reduces soil C inputs from root exudates and leaf litters by decreasing plant root and leaf biomass (Figure 1A). In addition, the physiological stress caused by e[O_3] might directly inhibit soil microbial activities,[18,49] as evidenced by the decreases in soil microbial biomass (phospholipid-derived fatty acids, −12.2%) and the activities of C-degrading (ligninase to cellulase) enzymes (10.4%–28.0%) (Figure 2G; Table S7). This reduction in microbial activity and the associated microbial respiration may partly explain the decrease in soil respiration under e[O_3] (10.4%) (Figure 2J),[8,50] along with the determinant cause of decreased litter inputs and root biomass (root respiration). However, e[O_3] reduces soil C inputs more strongly than soil respiration, which is consistent with a decrease in SOC and soil total N (TN) contents by 5.8% and 6.2% (Figure 2G), respectively, and a slight increase in soil C/N ratio by 1.3% (Figure S2). However, the responses of soil C and N pools to e[O_3] vary with ecosystem type and e[O_3] duration (see Note S2). Overall, on a global scale, e[O_3] could potentially reduce SOC storage and TNP (0–20 cm layer) by 22.3 (95% CI, 7.5–37.2) Pg CO_2-eq year^{-1} and 0.08 (95% CI, −0.35 to 0.19) Pg N year^{-1}, respectively (Figure 6), suggesting that e[O_3] largely damages the capacity of terrestrial ecosystems to store C and N in soils.

The various soil labile C and N pools also decreased under e[O_3], but to a greater extent than SOC and TN (dissolved organic C [DOC], 15.2%; microbial biomass C [MBC], 15.5%; microbial biomass N [MBN], 22.9%; and NH_4^+, 16.4%) (Figure 2). Soil labile C and N serve as metabolized substrates or energy sources for microbial organisms to produce GHGs (see also Note S5).[25,28] For example, methanogenic archaea utilize organic C substrates (e.g., DOC) under anaerobic conditions to produce CH_4,[19] while NH_4^+ and NO_3^- are the primary N substrates for

Apart from [O_3] concentration, climate conditions and ecosystem types also regulate the responses of plant NPP and N pools to e[O_3] (see Note S1 for details).[5] Decreases in plant NPP (ANPP and BNPP) and (leaf) N pools with e[O_3] were significantly and positively correlated with mean annual temperature (MAT) and precipitation (MAP), suggesting that the plant productivity of subtropical and tropical terrestrial ecosystems may be hit hardest by elevated [O_3]. For ecosystems in cold and arid regions, plant photosynthesis and growth are relatively less likely to be inhibited by e[O_3] because the physiological stresses from low temperature and water deficiency make plants less susceptible to the additional e[O_3] stress.[43,44] In contrast, when e[O_3] becomes the primary stress in subtropical and tropical regions, reductions in plant photosynthesis and productivity are more severe.[4,5] These findings corroborate simulations of the impact of e[O_3] on global gross primary production (GPP), which showed that decreases in GPP in equatorial regions with higher MAT and MAP were greater than in North America and Asia.[42] Thus, to minimize the damages of e[O_3] on global C and N storages, there is an urgent need for air pollution control specifically in subtropical and tropical regions from the Middle East to India and East Asia where tropospheric O_3 concentrations have strongly increased in recent decades.[45]

lnRR of variables	MAT	MAP	Clay$_{content}$	SOC$_{content}$	TN$_{content}$	[O$_3$]$_{lnRR}$	[O$_3$]$_{hours/day}$
Aboveground bimass	0.02	0.03	0.15	-0.21	-0.06	-0.21**	-0.11
Root bimass	-0.21**	-0.22**	-0.08	0.02	-0.03	-0.34***	-0.14*
Leaf bimass	0.01	-0.11	-0.19	-0.04	0.53**	0.03	-0.07
Stem bimass	-0.08	0.01	0.56**	-0.49*	0.12	0.05	-0.03
Crop yield	0.05	-0.01	0.11	-0.16	-0.01	-0.61***	0.11
P_n	-0.15*	0.05	-0.22	0.11	0.05	-0.21**	0.03
g_s	-0.04	0.04	0.37	0.34	-0.08	-0.25**	-0.25**
WUE	0.06	-0.08	0.22	0.09	0.11	-0.24	0.12
v_{cmax}	-0.21	-0.05	0.05	-0.15	-0.25	0.16	-0.06
Chlorophyll	-0.22*	-0.41***	0.64**	-0.57*	0.07	-0.44***	0.16
ANPP	-0.65**	-0.52*	-0.08	0.01	-0.03	-0.72***	0.26
BNPP	-0.38*	-0.49*	-0.25	0.01	0.01	-0.68***	-0.17
Leaf C pool	-0.51*	-0.56**	-0.08	0.03	-0.12	-0.57**	-0.03
Stem C pool	-0.08	-0.03	0.01	0.01	0.03	-0.18	-0.11
Yield C pool	-0.22	-0.11	-0.08	0.02	-0.03	-0.15	-0.15
Aboveground N pool	-0.08	-0.12	0.18	-0.08	-0.14	-0.25*	0.03
Belowground N pool	-0.05	-0.12	-0.35	0.68***	0.57**	-0.27*	0.01
Leaf N pool	-0.29*	-0.26*	0.51*	-0.14	0.61***	0.1	0.07
Stem N pool	-0.43**	-0.17	0.21	-0.12	0.54**	0.26	0.01
Yield N pool	-0.31*	-0.21	-0.12	-0.11	0.09	-0.51***	0.18
SOC content	-0.12	-0.11	-0.13	0.23	0.35	0.06	-0.07
DOC content	-0.66***	0.55***	-0.35	-0.08	0.01	-0.09	-0.31
MBC	0.08	-0.13	0.01	0.19	0.05	-0.52**	0.29
TN content	0.04	0.08	-0.12	-0.18	0.01	-0.05	-0.34*
MBN	-0.31	-0.55**	-0.11	-0.41	0.27	0.01	-0.06
Ecosystem respiration	-0.11	-0.42	-0.85***	-0.31***	0.24	-0.44**	0.24
Soil respiration	-0.13	-0.39*	-0.77***	-0.28	-0.36	-0.38**	0.32
CH$_4$ emissions	-0.48*	0.07	-0.55	0.53	0.42	-0.39*	0.21
N$_2$O emissions	-0.31	-0.3	0.11	-0.05	0.07	-0.31*	0.11

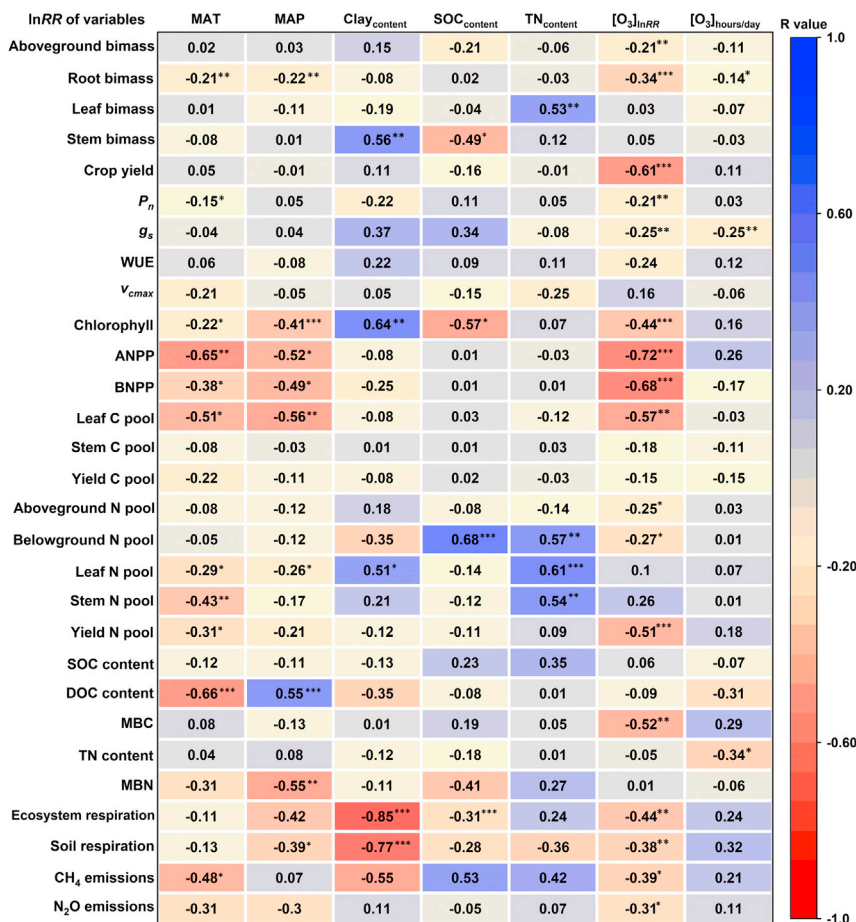R value scale: 1.0 — 0.60 — 0.20 — -0.20 — -0.60 — -1.0

Figure 3. Environmental and edaphic factors regulating e[O$_3$] impacts on C and N pools and fluxes

Linear regression analysis between terrestrial ecosystems C and N pools and fluxes with climate conditions, initial soil properties, and elevated O$_3$ conditions. The lnRR of variables represents the response ratio of a given variable to elevated O$_3$. Clay$_{content}$, initial soil clay content; MAP, mean annual precipitation; [O$_3$]$_{hours/day}$, elevated O$_3$ running hours/day in the treatment plot; [O$_3$]$_{lnRR}$, natural log of the response ratio (RR) of elevated O$_3$ concentration/ambient O$_3$ concentration; P_n, net photosynthetic rate; SOC$_{content}$, initial SOC content; V_{cmax}, the maximum carboxylation rate; WUE, water use efficiency. Negative (positive) R value denotes a negative (positive) linear relationship (*0.01 < p < 0.05, **0.001 < p < 0.01, ***p < 0.001).

N$_2$O production through heterotrophic nitrification and denitrification,[51] with DOC acting as an energy source.[52] Across all studies, the decreased availability of soil inorganic N and DOC under e[O$_3$] significantly reduced the rates of soil nitrification (−18.0%) and denitrification (−9.2%) (Figure 4; Tables S8 and S9), thereby decreasing N$_2$O emissions (−12.1%, equivalent to −1.36 Pg CO$_2$-eq year^{-1}; Figures 2J, 4, and S12). CH$_4$ emissions from paddy fields and wetlands were also decreased by 19.2% (−0.57 Pg CO$_2$-eq year^{-1}) (Figures 2J, 6, and S3), likely owing to a lower supply of organic C substrates.[19,20] Previous studies postulated that e[O$_3$]-induced reductions in soil CH$_4$ and N$_2$O emissions may negate the decreases in terrestrial net ecosystem production (NEP) and SOC storage.[5,16,19] However, according to our analysis this was unlikely, as the decreases in soil CH$_4$ and N$_2$O emissions (−1.9 Pg CO$_2$-eq year^{-1}) under e[O$_3$] were smaller than the decreases in NEP (−3 Pg CO$_2$-eq year^{-1}) and SOC pool (−22.3 Pg CO$_2$-eq year^{-1}) (Figure 6). As a result, the overall source strength of global terrestrial ecosystems for GHGs under e[O$_3$] was increased by 20.4 Pg CO$_2$-eq year^{-1} (nGWP).

e[CO$_2$] negates e[O$_3$] impacts on terrestrial C and N cycles

In contrast with e[O$_3$], e[CO$_2$] generally increases plant photosynthesis and growth.[24,27,29] Averaged across all studies in our dataset, e[CO$_2$] significantly stimulated plant photosynthesis by 32.6% (Figure 1E) and various plant tissue biomass by 19.4%–33.7% in global terrestrial ecosystems (Figure 1B). Plants exposed to e[CO$_2$] showed reduced g_s (13.0%) (Figure 1E), which decreases O$_3$ flux into plants and consequently its detrimental effects on plant photosynthesis.[5,41,53] Our results showed that e[O$_3$] × e[CO$_2$] largely alleviated the decrease in V_{cmax} and leaf Chll under e[O$_3$] and significantly stimulated photosynthetic WUE (Figure 1F). As a result, plant photosynthesis was significantly increased by 16.7% under e[O$_3$] × e[CO$_2$], leading to an overall increase in the biomass of plant tissues (12.8%–20.8%) and crop yield (7.4%) (Figures 1C and S5). Also, the root/shoot ratio under e[O$_3$] × e[CO$_2$] remained unchanged compared with ambient conditions (Figure S2), which suggests that e[CO$_2$] enhanced the tolerance of plants to e[O$_3$] damage.[5,53] As the stimulation in plant P_n increased significantly with the level of [CO$_2$] enrichment (Tables S1 and S10), the increases in e[CO$_2$] can overcome the decreased terrestrial plant productivity under e[O$_3$].

Plant NPP and its components (ANPP, BNPP, leaf, and stem C pools) were significantly increased by 19.8%–31.6% under e[CO$_2$] and by 17.3%–33.2% under e[O$_3$] × e[CO$_2$] (Figures 2B, 2C, and S9), because of the enhanced plant biomass and unaffected or stimulated plant C concentrations (Figure 1). In contrast to e[O$_3$], e[CO$_2$] and e[O$_3$] × e[CO$_2$] decreased plant N concentrations (Figures 1K and 1L), also reported in other studies,[23,48,54] which resulted in an increase of plant C/N ratios (Figure S2). The negative effects of e[CO$_2$] on plant N concentrations were likely due to the growth dilution effect.[54] The decrease in plant N concentrations was comparable in magnitude with the increase in plant biomass under e[O$_3$] × e[CO$_2$] and led to a slight but nonsignificant increase in various plant N pools (2.1%–10.7%). These N pools were significantly increased (5.4%–17.1%) under e[CO$_2$] alone (Figures 2E and 2F). Regarding the interactive effects of e[O$_3$] and e[CO$_2$], additive effects were observed for
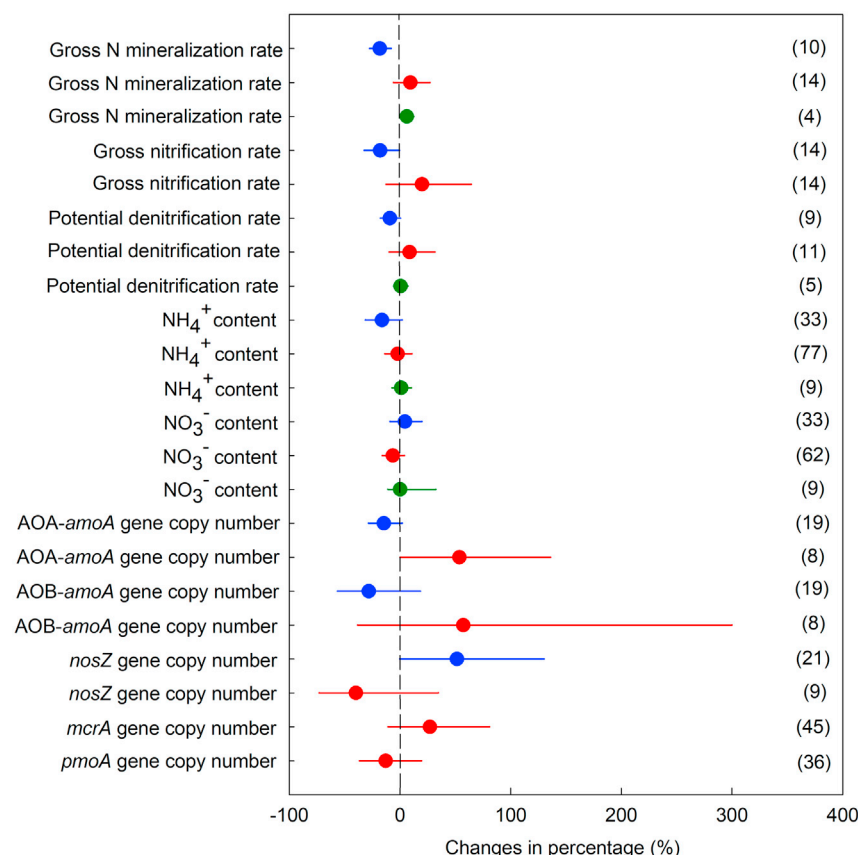
Figure 4. Responses of factors regulating N₂O and CH₄ emissions to e[O₃], e[CO₂], and e[O₃] × e[CO₂]

Changes in soil N transformation rates, soil N availability, and the functional gene copy number associated with N_2O and CH_4 emissions under elevated O_3 (e[O_3], on average 72 ppbv, blue color), elevated CO_2 (e[CO_2], on average 615 ppmv, red color), and e[O_3] (on average 67 ppbv) × e[CO_2] (on average 612 ppmv, dark green color) in terrestrial ecosystems. The number of experimental observations is in parentheses.

eq year^{-1} and 0.011 Pg N year^{-1} under e[CO_2] and e[O_3], respectively (Figure 6).

The e[CO_2]-induced increase in soil organic and labile C and N pools are known to promote soil GHG emissions.[25,54,55] We found that, on average, e[CO_2] increased soil emissions of CO_2 by 29.0% (equivalent to 23.7 Pg CO_2-eq year^{-1}), CH_4 by 31.8% (1.1 Pg CO_2-eq year^{-1}) (paddy fields and wetlands), and N_2O by 16.4% (1.5 Pg CO_2-eq year^{-1}) (Figures 2K and 6). Partially offset by the negative effects of e[O_3] (Figure 2L), the increases in soil CO_2 (2.6 Pg CO_2-eq year^{-1}), CH_4 (0.50 Pg CO_2-eq year^{-1}), and N_2O (0.15 Pg CO_2-eq year^{-1}) emissions under e[CO_2] and e[O_3] were lower than e[CO_2] only (Figure 6). Our analyses suggest that simultaneous increase in [CO_2] and [O_3] significantly diminishes the expected increase in the sink strength of terrestrial ecosystems for GHGs under e[CO_2] alone by 81.5%. Nevertheless, global terrestrial ecosystems remain a C sink of about 5.7 Pg CO_2-eq year^{-1} (nGWP) under e[CO_2] and e[O_3], largely owing to the increases in SOC storage (6.3 Pg CO_2-eq year^{-1}), which are not fully compensated by the increases in soil CH_4 and N_2O emissions. These results suggest that the negative effects of e[O_3] on terrestrial ecosystems are overestimated if e[CO_2] impacts are not considered, stressing the need for revising current global CN-models regarding the parameterization of e[O_3] and e[CO_2] interactive effects.

Summary and the way forward

Our meta-analysis showed that, compared with ambient concentrations, elevated [O_3] significantly decreased plant productivity, and plant and soil C and N pools in global terrestrial ecosystems (Figure 7). However, the reductions in these C and N fluxes will be offset by the positive effects of e[CO_2] on ecosystem C and N cycling, since the interactive effects of e[O_3] and e[CO_2] are mostly additive (Figure 5). Regardless of the levels of [O_3] and [CO_2] enrichment, elevated CO_2 negates O_3 impacts on terrestrial C and N cycles (Figures S20−S24; Note S3). These results did not necessarily indicate that e[CO_2] is beneficial to ecosystem sustainability. Instead, we highlight that the complex interactive impacts of e[O_3] and e[CO_2] need to be considered when implementing
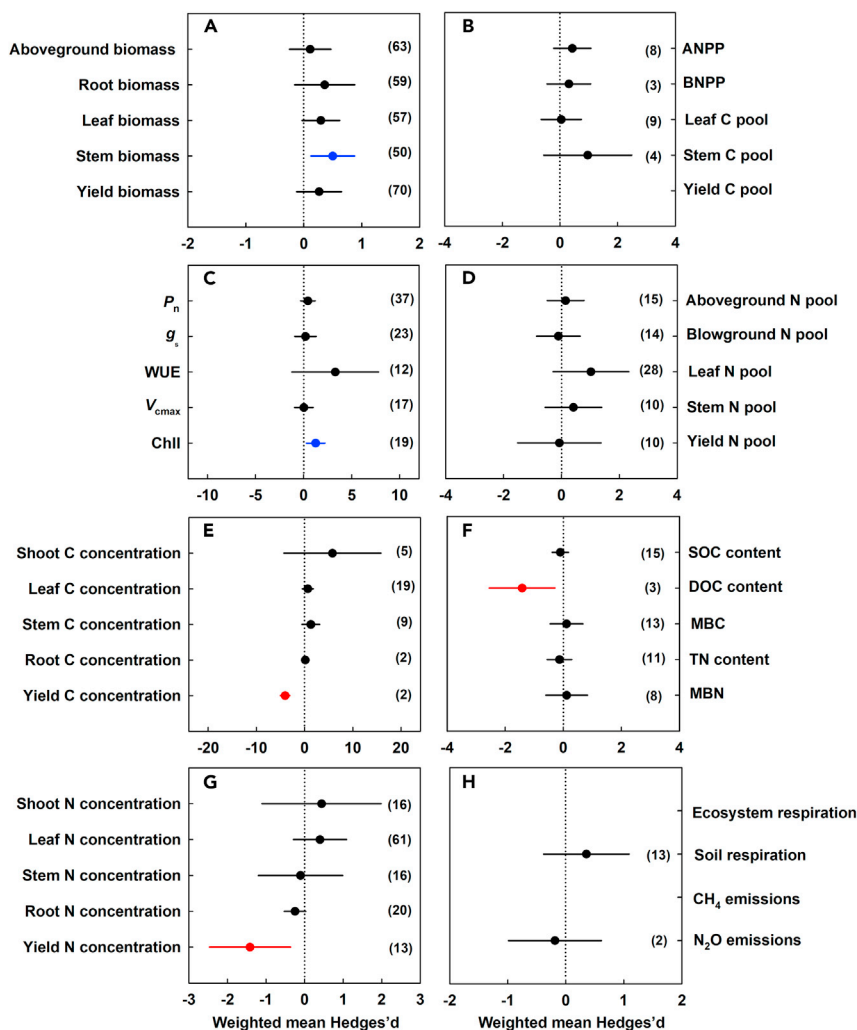
plant C and N pools (Figure 5). On a global scale, the increases in NPP and TNP of terrestrial ecosystems under e[CO_2] (31.3 Pg CO_2-eq year^{-1} and 0.37 Pg N year^{-1}) exceed the decreases induced by e[O_3] (−24.1 Pg CO_2-eq year^{-1} and −0.18 Pg N year^{-1}). Thus, we expect that the combined effect of e[O_3] and e[CO_2] is positive, i.e., will result in a net gain in terrestrial plant NPP and N pools by 7.1 Pg CO_2-eq year^{-1} and 0.19 Pg N year^{-1}, respectively (Figure 6). These results further strengthen our argument that e[CO_2] alleviates or even overcompensates the negative effects of e[O_3] on global terrestrial ecosystems (Figure 7).

The increase in plant biomass under e[CO_2] stimulates soil belowground C and N processes.[31,48] Our results showed that SOC and TN contents were slightly increased by 0.3% and 1.2% under e[O_3] × e[CO_2] (Figure 2G), mainly due to the enhanced soil C inputs through root and leaf litters.[28] These percentage changes are smaller than those observed under e[CO_2] alone (6.8% for SOC and 5.7% for TN) (Figure 2H). The difference was attributed to e[O_3] offsetting the positive effect of e[CO_2] on soil C and N pools.[8] Soil labile C (MBC and DOC) and N (MBN and NH_4^+) contents were also slightly stimulated, respectively, by 0.2%–2.2% and 1.3%–4.3% under e[O_3] × e[CO_2], although these effects were not significant (Figure 2F). The positive effects of e[CO_2] on soil C and N pools may become more evident under long-term [CO_2] elevation (Figure S4).[15] Globally, the increases in SOC storage and TNP (0–20 cm layer) under e[CO_2] outweigh the expected negative effects by e[O_3], resulting in a net increase of the soil C and N stocks in terrestrial ecosystems by 6.3 Pg CO_2-

Figure 5. Interactions between e[O₃] and e[CO₂] on C and N pools and fluxes

Interactive effects of elevated O_3 (e[O_3]) and elevated CO_2 (e[CO_2]) on plant biomass (A), plant NPP (B), plant photosynthesis (C), plant N pools (D), plant C concentrations (E), soil C and N pools (F), plant N concentrations (G), and greenhouse gas emissions (H) in global terrestrial ecosystems. The number of experimental observations is in parentheses. Weighted mean Hedges' d values labeled with black, blue, and red represent the interactive effects between e[O_3] and e[CO_2] are additive, antagonistic, and synergistic, respectively.

mitigation measures to curb the rising levels of CO_2 or O_3. Our findings further emphasize the importance of exploring the interactive effects of multiple global change drivers, to better understand how changes in the global atmospheric composition will affect the C and N cycles of terrestrial ecosystems. Moreover, the interactive effects of other global change factors (e.g., warming and drought) should also be explored, and the likely complicated responses of ecosystem processes to dynamically changing environmental factors need to be studied by well-designed multifactorial experiments,[31,56,57] to develop a more realistic view on future ecosystem functioning.

Most studies in our dataset were conducted in temperate regions, although the countries in the subtropics and tropics might be hit hardest by e[O_3][42] and are more vulnerable to environmental changes owing to the importance of agriculture to their economies. Thus, there is an urgent need for more e[O_3] experiments in tropical regions. Overall, our study provides new insights into improving the prediction of C and N models in capturing the complex responses of terrestrial ecosystems to future atmospheric conditions.

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, Dr. Longlong Xia (longlong.xia2@kit.edu).

Materials availability
This study did not generate new unique materials.

Data and code availability
The datasets generated during this study are available at https://doi.org/10.17632/4xft935wvy.1.

Data compilation
Peer-reviewed publications before July 2020 that reported the effects of e[O_3], e[CO_2], and e[O_3] × e[CO_2] on terrestrial C and N cycles were collected using the databases such as Web of Science, Scopus, Google Scholar, CAB Abstracts, Academic Search Complete (EBSCO), Current Contents Connect (ISI), China National Knowledge Infrastructure Database (CNKI), and China Wanfang Database. The keywords used in the search included "elevated O_3, elevated CO_2, or elevated O_3 and CO_2," "plant photosynthesis (net P_n, g_s, water use efficiency (WUE), the maximum carboxylation rate (V_{cmax}) and leaf chlorophyll concentration (Chll))," "plant and its above- and belowground components biomass (shoot, root, leaf, and stem)," "root/shoot ratio," "food
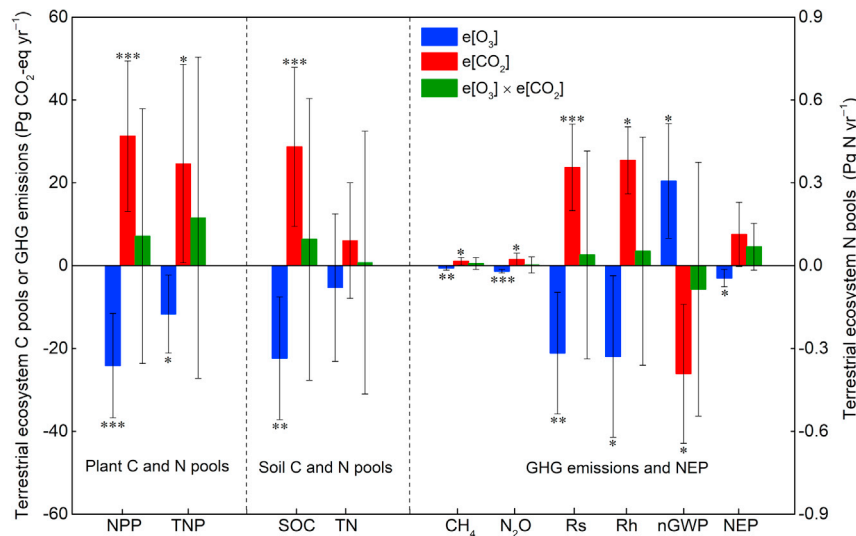
Figure 6. Net changes in global terrestrial ecosystem C and N pools and fluxes in response to e[O₃], e[CO₂], and e[O₃] × e[CO₂]
All C pools and GHG emissions were converted to CO_2-eq. CO_2-eq emissions were calculated using the 100-year global warming potentials of 34 and 298 for CH_4 and N_2O, respectively. CH_4 emissions represent the changed emissions from global paddy fields and wetlands; nGWP (net global warming potential) = CH_4 + N_2O – SOC, positive nGWP denotes the increased source strength due to changes in atmospheric composition; NEP = NPP – Rh; NPP, net primary productivity; Rh, soil heterotrophic respiration; Rs, soil respiration; SOC, SOC pool (0–20 cm layer); TN, soil TNP (0–20 cm layer); TNP, plant TN pool. The asterisks represent the changes are significant (*0.01 < p < 0.05, **0.001 < p < 0.01, ***p < 0.001). The error bars represent the 95% CIs. Since the interactive effects of e[O₃] and e[CO₂] on plant, soil C and N pools, and GHG emissions were additive (Figure 5), the changes for various C and N variables under simultaneous e[O₃] and e[CO₂] were calculated by summing the values for e[O₃] alone and e[CO₂] alone (i.e., e[O₃] and e[CO₂]ᵥₐᵣᵢₐᵦₗₑ = e[O₃]ᵥₐᵣᵢₐᵦₗₑ + e[CO₂]ᵥₐᵣᵢₐᵦₗₑ).

security (crop yield)," "plant and its components C concentrations," "plant NPP and its components C pools," "plant and its components N concentrations," "plant and its components N pools," "plant C/N ratio (shoot C/N ratio, root C/N ratio, leaf C/N ratio and stem C/N ratio)" "soil C and N pool and their ratios (0–20 cm layer) (SOC and TN contents, and soil C/N ratio)," "soil labile C and N pools (DOC, microbial C and N biomass and their ratios (MBC and MBN, and MBC/MBN), and ammonium (NH_4^+) and nitrate (NO_3^-) contents)," "GHG emissions (ecosystem and soil respiration, and CH_4 and N_2O emissions)," "N_2O-related N transformation rates (mineralization, nitrification, and denitrification)," "N_2O-related functional gene (AOA-*amoA*, AOB-*amoA* and nosZ) copies" and "CH_4-related functional gene (mcrA and pmoA) copies." Studies that met the following criteria were included in this meta-analysis: (a) at least one of the target variables for the control (ambient concentration) and treatment (e[O₃], e[CO₂], or e[O₃] × e[CO₂]) plots was included; (b) the experimental duration and e[O₃], e[CO₂], or e[O₃] × e[CO₂] fumigation details (e.g., fumigation time and duration) were reported; and (c) the mean values and sample sizes for the control and treatment plots were provided. The multiple observations that were conducted at the same experimental site over several years were averaged. Besides, to better represent the canopy effects, we averaged the measurements of photosynthetic traits (P_n, g_s, V_{cmax}, and Chll) of leaves located in different positions of the canopy when available. In total, 810 peer-reviewed publications (9,109 observations) reporting results from global terrestrial ecosystems, mainly grassland (123 studies), forest (246 studies), and croplands (408 studies), were included in the meta-analysis (Figure S1).

Meta-analysis
The natural log-transformed response ratio (ln*RR*) was used to quantify the effects of e[O₃], e[CO₂], or e[O₃] × e[CO₂] on the variables (X) using the following equation:

$$\ln RR = \ln (X_t / X_c) \qquad \text{(Equation 1)}$$

where X_t and X_c represent the mean of the treatment (e[O₃], e[CO₂], or e[O₃] × e[CO₂]) and control groups (ambient concentration) for the variable X, respectively. The results are presented as the percentage of changes ((RR – 1) × 100) in the variables under e[O₃], e[CO₂], or e[O₃] × e[CO₂]. Positive percentage changes denote an increase due to e[O₃], e[CO₂], or e[O₃] × e[CO₂], whereas negative values indicate a decrease in the respective variables. The effect size was weighted by the inverse of its variance and the missing variance was estimated using the average coefficient of variation across the dataset.[58] We

performed a mixed-effects meta-analysis in R, using the rma.mv function in "metaphor" package which included "paper" as a random effect.[59]

The effects of e[O₃], e[CO₂], or e[O₃] × e[CO₂] were considered significant if the 95% CIs did not overlap with zero. The means of the categorical variables (ecosystem types and fumigation methods) were considered significantly different from each other if their 95% CIs did not overlap. For each variable, total heterogeneity among the categorical group (Q_t) was partitioned into within-group (Q_w) and between-group (Q_b). A significance of Q_b (p < 0.05) represents a significant difference between different levels of the categorical group (Tables S4 and S5). Meta-regression was conducted between ln*RR* of the target variables and climate parameters (MAT and MAP), soil properties (SOC and TN contents and clay content), and [O₃] and [CO₂] concentrations (Figure 3; Tables S1 and S2). Publication bias for the variables included in this meta-analysis was assessed using funnel plots.

Interactive effects
The interactive effects of e[O₃] and e[CO₂] on the variables were evaluated using Hedges' d. The interactive effect size (d_I) between e[O₃] and e[CO₂] was calculated using the following equation[27,60]:

$$d_I = \frac{(X_{AB} - X_A) - (X_B - X_C)}{2s} \times J(m) \qquad \text{(Equation 2)}$$

where X_C, X_A, X_B, and X_{AB} denote the mean of a variable in the control, e[O₃], e[CO₂], and their combination (e[O₃] × e[CO₂]), respectively; s and $J(m)$ denote the pooled standard deviation and correction term for small sample bias, respectively, which were calculated using the following equations:

$$s = \sqrt{\frac{(n_c - 1)s_c^2 + (n_A - 1)s_A^2 + (n_B - 1)s_B^2 + (n_{AB} - 1)s_{AB}^2}{n_c + n_A + n_B + n_{AB} - 4}} \qquad \text{(Equation 3)}$$

$$J(m) = 1 - \frac{3}{4m - 1} \qquad \text{(Equation 4)}$$

where n_c, n_A, n_B, and n_{AB} denote the sample size, and s_c, s_A, s_B, and s_{AB} represent the standard deviation in the control e[O₃], e[CO₂], and their combination (e[O₃] × e[CO₂]), respectively; m is the degree of freedom ($m = n_c + n_A + n_B + n_{AB} - 4$). We calculated the variance of d_I (v) using the following equation:

$$v = \frac{1}{4}\left[\frac{1}{n_c} + \frac{1}{n_A} + \frac{1}{n_B} + \frac{1}{n_{AB}} + \frac{d_I^2}{2(n_c + n_A + n_B + n_{AB})}\right] \qquad \text{(Equation 5)}$$
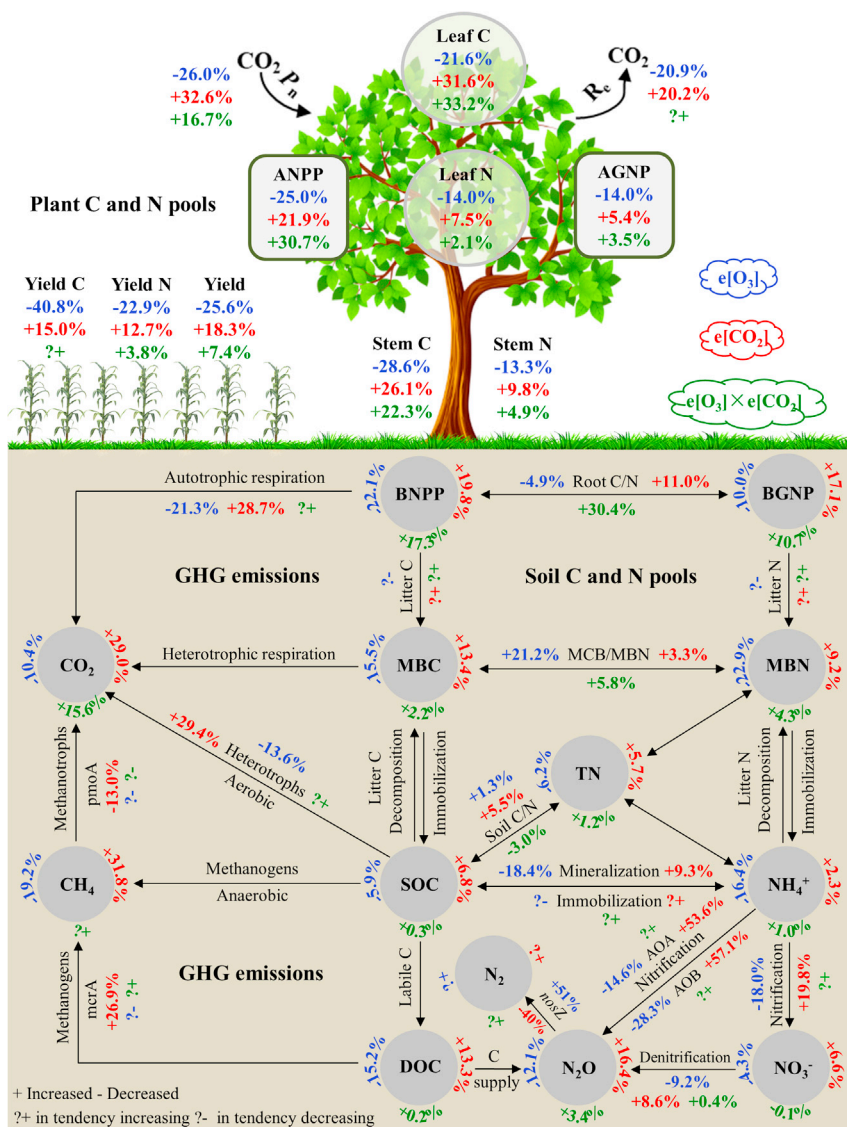
Figure 7 (conceptual model)

Plant C and N pools

CO_2 P_n
-26.0%
+32.6%
+16.7%

Leaf C
-21.6%
+31.6%
+33.2%

CO_2 R_e
-20.9%
+20.2%
?+

ANPP
-25.0%
+21.9%
+30.7%

Leaf N
-14.0%
+7.5%
+2.1%

AGNP
-14.0%
+5.4%
+3.5%

Yield C
-40.8%
+15.0%
?+

Yield N
-22.9%
+12.7%
+3.8%

Yield
-25.6%
+18.3%
+7.4%

e[O_3]
e[CO_2]
e[O_3] × e[CO_2]

Stem C
-28.6%
+26.1%
+22.3%

Stem N
-13.3%
+9.8%
+4.9%

Autotrophic respiration
-21.3% +28.7% ?+

BNPP ×22.1% +19.8% ×17.5%

-4.9% Root C/N +11.0%
+30.4%

BGNP ×10.0% +17.1% ×10.7%

GHG emissions

Soil C and N pools

Litter C ?- ?+ ×15.5%

Litter N ?- ?+ ×22.9%

Heterotrophic respiration
CO_2 ×10.4% +29.0% ×15.6%

MBC +21.2% MCB/MBN +3.3% +5.8% ×13.4% ×2.2%

MBN ×9.2% ×4.3%

+29.4% Heterotrophs ?+
-13.6% Aerobic

Methanotrophs pmoA
-13.0% ?- ?-

Litter C Decomposition Immobilization

Litter N Decomposition Immobilization

TN +1.3% +5.5% Soil C/N -3.0% -6.2% +5.7% ×1.2%

CH_4 ×19.2% +31.8%

Methanogens Anaerobic

SOC -5.9% +6.8% ×0.3%

-18.4% Mineralization +9.3%
?- Immobilization ?+
?+

NH_4^+ -16.4% +2.3% ×1.0%

Methanogens mcrA
+26.9% ?- ?+ ?-

GHG emissions

Labile C ?+ ?-

N_2 ?- ?+

-14.6% AOA +53.6% ?+
Nitrification
-28.3% AOB +57.1% ?+

Nitrification -18.0% +19.8% ?+

NO_3^- +6.6% ×0.1%

DOC ×15.2% +13.3% ×0.2%

C supply -12.1% +51% nosZ ?+ -40%

N_2O +16.4% ×3.4%

Denitrification -9.2% +8.6% +0.4%

+ Increased − Decreased
?+ in tendency increasing ?- in tendency decreasing

Figure 7. A conceptual model summarizing terrestrial C and N feedbacks to e[O_3], e[CO_2] and e[O_3] × e[CO_2]

Overall effects of elevated [O_3] (blue), elevated [CO_2] (red), and e[O_3] × e[CO_2] (dark green) on plant C and N pools, soil C and N pools, and greenhouse gas emissions in terrestrial ecosystems. ANPP, aboveground net primary productivity; BNPP, belowground net primary productivity; AGNP, aboveground N pool; BGNP, belowground N pool; SOC, soil organic C; DOC, dissolved organic C; MBC, microbial biomass C; MBN, microbial biomass N.

The weighted mean d_l (d_{++}) was calculated using the following equation:

$$d_{++} = \frac{\sum_{i=1}^{l}\sum_{j=1}^{k} w_{ij}d_{ij}}{\sum_{i=1}^{l}\sum_{j=1}^{k} w_{ij}} \qquad \text{(Equation 6)}$$

where l represents the group number, k is the number of comparisons in the ith group, and w is the weight that is the reciprocal of the variance ($1/v$). Weighted mean d_l and the 95% CIs were calculated using the "metafor" package in R software.[59] The interactive effects of e[O_3] and e[CO_2] on the variables were additive, synergistic, or antagonistic.[60] The interactive effect was considered to be additive if the 95% CI overlapped with zero. For two-driver pairs whose individual effects were either both negative or had opposite directions, the interactive effect sizes less than zero were synergistic whereas those greater than zero were antagonistic.

Upscaling and uncertainty analysis

We scaled up the weighted and averaged results (area-scaled metrics, $\bar{U}$-value) from this meta-analysis by multiplying them for target variables with the corresponding total land area[25,28]:

$$T = \bar{U} \times A \qquad \text{(Equation 7)}$$

where T is expressed in Pg CO_2-eq year^{-1} or Pg N year^{-1} and A is the total land area for wetlands (5.7 million km^2), rice paddies (1.3 million km^2), and upland (103 million km^2).[25,28] When scaling up N_2O emissions, we distinguished between fertilized upland soils (i.e., 19 million km^2 of fertilized grasslands and croplands minus 1.3 million km^2 rice paddies) and nonfertilized upland soils (85.4 million km^2).[28] U was weighted by the inverse of its variance. CO_2-eq emissions were calculated using the 100-year global warming potentials of 34 and 298 for CH_4 and N_2O, respectively.[1]

For paired data, we determined the SOC and TN change rate (MU, g C, or N kg^{-1} year^{-1}) under atmospheric composition changes by plotting them versus the experimental duration, which was defined and calculated using the following equation:[28]

$$\text{MU} = (\text{SOC}_t\,(\text{TN}_t) - \text{SOC}_c\,(\text{TN}_c))/t \qquad \text{(Equation 8)}$$

where SOC_t (TN_t) and SOC_c (TN_c) represent the mean SOC (TN) change rate in treatment (e[O_3], e[CO_2] or e[O_3] × e[CO_2]) and control groups (ambient concentration), respectively. Furthermore, the SOC and TN pool (SOC$_p$ and TN$_p$, t C or N year^{-1}) change rate (0–20 cm layer) was estimated using the following equation[61,62]:

$$SOC_p\ (TN_p) = \sum_{i=1}^{m} A_i \times MU \times BD \times H \times 0.1 \qquad \text{(Equation 9)}$$

where A_i is the habitat area (i.e., upland, wetland, and paddy field); BD is the bulk density (g cm^{-3}) and H is the depth of the soil layer (20 cm). Since the interactive effects of e[O$_3$] and e[CO$_2$] on plant and soil C and N pools, and GHG emissions were additive (Figure 6), the changes for various C and N variables under simultaneous e[O$_3$] and e[CO$_2$] were calculated by summing the values for e[O$_3$] alone and e[CO$_2$] alone (i.e., e[O$_3$] and e[CO$_2$]$_{variable}$ = e[O$_3$]$_{variable}$ + e[CO$_2$]$_{variable}$) (Figure 6).

Uncertainties exist in our estimation of changes in C and N pools and GHG emissions on a global scale. First, the responses of GHG emissions, and soil C and N pools to e[O$_3$] and e[CO$_2$] may change over the experimental duration (Figure S4), which was not considered in the current estimation due to data limitation for long-term studies. Second, we assumed that the effects of e[O$_3$] and e[CO$_2$] mainly occur during the plant-growing season when converting seasonal fluxes to annual data, which may affect the upscaling results.[25] Third, the responses of soil C and N pools and GHG emissions to e[O$_3$] and e[CO$_2$] may vary with climate conditions (e.g., temperature and precipitation) and soil properties (e.g., initial SOC content) (Figure 3 and S13), which was not assessed in the analysis. Fourth, the effects of O$_3$ are dose dependent and plants often exhibit threshold responses to cumulative doses of O$_3$ (Figures S14−S19). For example, staple crop species (e.g., rice, wheat, and corn) are less tolerant to e[O$_3$] than tree species (e.g., spruce, silver fir, and pine) based on the AOT$_{40}$ index values (accumulated exposure over threshold of 40 ppbv) (Table S3). Our results further show that e[O$_3$] levels of around 70 ppbv in combination with e[CO$_2$] levels of around 550 ppmv increased the aboveground biomass of food crops (e.g., wheat, bean, and vegetables) at a similar or even greater magnitude than for tree species (e.g., pine and spruce) being exposed to a combination of e[O$_3$] of about 70 ppbv and e[CO$_2$] of about 700 ppmv (Figure S15). These results indicate that food crops respond more dramatically than tree species to concurrently changing e[O$_3$] and e[CO$_2$] levels. However, due to a lack of data, especially on the responses of plant/ soil C and N pools to e[O$_3$] and e[CO$_2$] and the species effects (Data S7), we did not upscale the results for individual plant species (see Note S2 for details). This underscores the importance of the inclusion of these factors into future studies to reduce the uncertainties.

REFERENCES

1. Hartmann, D.L., Klein Tank, A.M.G., Rusticucci, M., Alexander, L.V., Brönnimann, S., and Charabi, Y. (2013). IPCC Climate Change 2013: The Physical Science Basis, T.F. Stocker, et al., eds. (Cambridge Univ. Press).

2. WMO (2019). WMO Greenhouse Gas Bulletin: The State of Greenhouse Gases in the Atmosphere Based on Global Observations through 2018 (World Meteorological Organization).

3. Emberson, L. (2020). Effects of ozone on agriculture, forests and grasslands. Philos. Trans. R. Soc. A Math. Phys. Eng. Sci. *378*, 20190327.

4. Ashmore, M.R. (2005). Assessing the future global impacts of ozone on vegetation. Plant Cell Environ. *28*, 949–964.

5. Ainsworth, E.A., Yendrek, C.R., Sitch, S., Collins, W.J., and Emberson, L.D. (2012). The effects of tropospheric ozone on net primary productivity and implications for climate change. Annu. Rev. Plant Biol. *63*, 637–661.

6. Wittig, V.E., Ainsworth, E.A., and Long, S.P. (2007). To what extent do current and projected increases in surface ozone affect photosynthesis and stomatal conductance of trees? A meta-analytic review of the last 3 decades of experiments. Plant Cell Environ. *30*, 1150–1162.

7. Wittig, V.E., Ainsworth, E.A., Naidu, S.L., Karnosky, D.F., and Long, S.P. (2009). Quantifying the impact of current and future tropospheric ozone on tree biomass, growth, physiology and biochemistry: a quantitative meta-analysis. Glob. Change Biol. *15*, 396–424.

8. Loya, W.M., Pregitzer, K.S., Karberg, N.J., King, J.S., and Giardina, C.P. (2003). Reduction of soil carbon formation by tropospheric ozone under increased carbon dioxide levels. Nature *425*, 705–707.

9. Zak, D.R., Kubiske, M.E., Pregitzer, K.S., and Burton, A.J. (2012). Atmospheric CO$_2$ and O$_3$ alter competition for soil nitrogen in developing forests. Glob. Change Biol. *18*, 1480–1488.

10. Zak, D.R., Pregitzer, K.S., Kubiske, M.E., and Burton, A.J. (2011). Forest productivity under elevated CO$_2$ and O$_3$: positive feedbacks to soil N cycling sustain decade-long net primary productivity enhancement by CO$_2$. Ecol. Lett. *14*, 1220–1226.

11. Sitch, S., Cox, P.M., Collins, W.J., and Huntingford, C. (2007). Indirect radiative forcing of climate change through ozone effects on the land-carbon sink. Nature *448*, 791–794.

12. Pregitzer, K.S., Burton, A.J., King, J.S., and Zak, D.R. (2008). Soil respiration, root biomass, and root turnover following long-term exposure of northern forests to elevated atmospheric CO$_2$ and tropospheric O$_3$. New Phytol. *180*, 153–161.

13. Volk, M., Obrist, D., Novak, K., Giger, R., Bassin, S., and Fuhrer, J. (2011). Subalpine grassland carbon dioxide fluxes indicate substantial carbon losses under increased nitrogen deposition, but not at elevated ozone concentration. Glob. Change Biol. *17*, 366–376.

14. Kuzyakov, Y., Horwath, W.R., Dorodnikov, M., and Blagodatskaya, E. (2019). Review and synthesis of the effects of elevated atmospheric CO$_2$ on soil processes: No changes in pools, but increased fluxes and accelerated cycles. Soil Biol. Biochem. *128*, 66–78.

15. Talhelm, A.F., Pregitzer, K.S., Kubiske, M.E., Zak, D.R., Campany, C.E., Burton, A.J., Dickson, R.E., Hendrey, G.R., Isebrands, J.G., Lewin, K.F., et al. (2014). Elevated carbon dioxide and ozone alter productivity and ecosystem carbon content in northern temperate forests. Glob. Change Biol. *20*, 2492–2504.

16. Hu, E., Yuan, Z.J., Zhang, H.X., Zhang, W.W., Wang, X.K., Jone, S.B., and Wang, N.N. (2018). Impact of elevated tropospheric ozone on soil C, N and microbial dynamics of winter wheat. Agric. Ecosyst. Environ. *253*, 166–176.

17. Chen, W., Zhang, L.L., Li, X.Y., Ye, R.Z., Li, Q., Zhu, J.G., Fang, N.N., Wang, L.L., Wu, Z.J., Horwath, W.R., et al. (2015). Elevated ozone increases nitrifying and denitrifying enzyme activities in the rhizosphere of wheat after 5 years of fumigation. Plant Soil *392*, 279–288.

18. Chen, Z., Wang, X., Yao, F., Zheng, F., and Feng, Z. (2010). Elevated ozone changed soil microbial community in a rice paddy. Soil Sci. Soc. Am. J. *74*, 829–837.

19. Tang, H., Liu, G., Zhu, J., and Kobayashi, K. (2015). Effects of elevated ozone concentration on CH_4 and N_2O emission from paddy soil under fully open-air field conditions. Glob. Change Biol. *21*, 1727–1736.

20. Toet, S., Ineson, P., Peacock, S., and Ashmore, M. (2011). Elevated ozone reduces methane emissions from peatland mesocosms. Glob. Change Biol. *17*, 288–296.

21. Kasurinen, A., Biasi, C., Holopainen, T., Rousi, M., Mäenpääm, M., and Oksanen, E. (2012). Interactive effects of elevated ozone and temperature on carbon allocation of silver birch (Betula pendula) genotypes in an open-air field exposure. Tree Physiol. *32*, 737–751.

22. King, J., Liu, L., and Aspinwall, M. (2013). Tree and forest responses to interacting elevated atmospheric CO_2 and tropospheric O_3: a synthesis of experimental evidence. In Developments in Environmental Science, *13*, R. Matyssek, N. Clarke, P. Cudlin, T.N. Mikkelsen, J.P. Tuovinen, G. Wieser, and E. Paoletti, eds. *(Elsevier), pp. 179–208*.

23. Ainsworth, E.A., and Long, S.P. (2005). What have we learned from 15 years of free-air CO_2 enrichment (FACE)? A meta-analytic review of the responses of photosynthesis, canopy properties and plant production to rising CO_2. New Phytol. *165*, 351–372.

24. Long, S.P., Ainsworth, E.A., Rogers, A., and Ort, D.R. (2004). Rising atmospheric carbon dioxide: plants FACE the future. Annu. Rev. Plant Biol. *55*, 591–628.

25. van Groenigen, K.J., Osenberg, C.W., and Hungate, B.A. (2011). Increased soil emissions of potent greenhouse gases under increased atmospheric CO_2. Nature *475*, 214–216.

26. van Groenigen, K.J., Qi, X., Osenberg, C.W., Luo, Y., and Hungate, B.A. (2014). Faster decomposition under increased atmospheric CO_2 limits soil carbon storage. Science *344*, 508–509.

27. Yue, K., Fornara, D.A., Yang, W.Q., Peng, C.H., Liu, Z.L., and Wu, F.Z. (2017). Influence of multiple global change drivers on terrestrial carbon storage: additive effects are common. Ecol. Lett. *20*, 663–672.

28. Liu, S.W., Cheng, J., Wang, C., Jin, Y.G., Zou, Z.H., Li, S.Q., Niu, S.L., and Zou, J.W. (2018). Climatic role of terrestrial ecosystem under elevated CO_2: a bottom-up greenhouse gases budget. Ecol. Lett. *21*, 1108–1118.

29. Luo, Y., Hui, D., and Zhang, D. (2006). Elevated CO_2 stimulates net accumulations of carbon and nitrogen in land ecosystems: a meta-analysis. Ecology *87*, 53–63.

30. Pregitzer, K.S., and Talhelm, A.F. (2013). Belowground carbon cycling at aspen FACE: dynamic responses to CO_2 and O_3 in developing forests. In Developments in Environmental Science, *13*, R. Matyssek, N. Clarke, P. Cudlin, T.N. Mikkelsen, J.P. Tuovinen, G. Wieser, and E. Paoletti, eds. *(Elsevier), pp. 209–226*.

31. Song, J., Wan, S.Q., Piao, S.L., Knapp, A.K., Classen, A.T., Vicca, S., Ciais, P., Hovenden, M.J., Leuzinger, S., Beier, C., et al. (2019). A meta-analysis of 1,119 manipulative experiments on terrestrial carbon-cycling responses to global change. Nat. Ecol. Evol. *3*, 1309–1320.

32. Malhi, Y., Franklin, J., Seddon, N., Solan, M., Turner, M.G., Field, C.B., and Knowlton, N. (2020). Climate change and ecosystems: threats, opportunities and solutions. Philos. Trans. R. Soc. B *375*, 20190104.

33. Morecroft, M.D., Duffield, S., Harley, M., Pearce-Higgins, J.W., Stevens, N., Watts, O., and Whitaker, J. (2019). Measuring the success of climate change adaptation and mitigation in terrestrial ecosystems. Science *366*, eaaw9256.

34. Grantz, D.A., Gunn, S., and Vu, H.B. (2006). O_3 impacts on plant development: a meta-analysis of root/shoot allocation and growth. Plant Cell Environ. *29*, 1193–1209.

35. Oikawa, S., and Ainsworth, E.A. (2016). Changes in leaf area, nitrogen content and canopy photosynthesis in soybean exposed to an ozone concentration gradient. Environ. Pollut. *215*, 347–355.

36. Ainsworth, E.A. (2008). Rice production in a changing climate: a meta-analysis of responses to elevated carbon dioxide and elevated ozone concentration. Glob. Change Biol. *14*, 1642–1650.

37. Feng, Z., Kobayashi, K., and Ainsworth, E.A. (2008). Impact of elevated ozone concentration on growth, physiology, and yield of wheat (*Triticum aestivum* L.): a meta-analysis. Glob. Change Biol. *14*, 2696–2708.

38. Mills, G., Sharps, K., Simpson, D., Pleijel, H., Frei, M., Burkey, K., Emberson, L., Uddling, J., Broberg, M., and Feng, Z.Z. (2018). Closing the global ozone yield gap: quantification and cobenefits for multistress tolerance. Glob. Change Biol. *24*, 4869–4893.

39. Zhu, X., Feng, Z.Z., Sun, T.F., Liu, X.C., Tang, H.Y., Zhu, J.G., Guo, W.S., and Kobayashi, K. (2011). Effects of elevated ozone concentration on yield of four Chinese cultivars of winter wheat under fully open-air field conditions. Glob. Change Biol. *17*, 2697–2706.

40. Cardoso-Vilhena, J., and Barnes, J. (2001). Does nitrogen supply affect the response of wheat (*Triticum aestivum* cv. Hanno) to the combination of elevated CO_2 and O_3? J. Exp. Bot. *52*, 1901–1911.

41. Valkama, E., Koricheva, J., and Okaanen, E. (2007). Effects of elevated O_3, alone and in combination with elevated CO_2, on tree leaf chemistry and insect herbivore performance: a meta-analysis. Glob. Change Biol. *13*, 184–201.

42. Soc, R. (2008). Ground-level Ozone in the 21st Century: Future Trends, Impacts and Policy Implications*15/08* (The Royal Society).

43. Smith, N.G., Keenan, T.F., Colin Prentice, I., Wang, H., Wright, I.J., Niinemets, Ü., Crous, K.Y., Domingues, T.F., Guerrieri, R., Yoko Ishida, F., et al. (2019). Global photosynthetic capacity is optimized to the environment. Ecol. Lett. *22*, 506–517.

44. Song, B., Niu, S., and Wan, S. (2016). Precipitation regulates plant gas exchange and its long-term response to climate change in a temperate grassland. J. Plant Ecol. *9*, 531–541.

45. Ziemke, J.R., Oamn, L.D., Strode, S.A., Douglass, A.R., Olsen, M.A., McPeters, R.D., Bhartia, P.K., Froidevaux, L., Labow, G.G., Witte, J.C., et al. (2019). Trends in global tropospheric ozone inferred from a composite record of TOMS/OMI/MLS/OMPS satellite measurements and the MERRA-2 GMI simulation. Atmos. Chem. Phys. *19*, 3257–3269.

46. Scharlemann, J.P.W., Tanner, E.V.J., Hiederer, R., and Kapos, V. (2014). Global soil carbon: understanding and managing the largest terrestrial carbon pool. Carbon Manag. *5*, 81–91.

47. Lal, R., Griffin, M., Apt, J., Lave, L., and Morgan, M.G. (2004). Managing soil carbon. Science *304*, 1623–1627.

48. Luo, Y., Sun, B., Currie, W.S., Dukes, J.S., Finzi, A., Hartwig, U., Hungate, B., McMurtrie, R.E., Oern, R., Parton, W.J., et al. (2004). Progressive nitrogen limitation of ecosystem responses to rising atmospheric carbon dioxide. BioScience *54*, 731–739.

49. Zhang, W., He, H.B., Li, Q., Lu, C.Y., Zhang, X.D., and Zhu, J.G. (2014). Soil microbial residue dynamics after 3-year elevated O_3 exposure are plant species-specific. Plant Soil *376*, 139–149.

50. Chen, J., Luo, Y., van Groenigen, K.J., Hungate, B., Cao, J., Zhou, X., and Wang, R. (2018). A keystone microbial enzyme for nitrogen control of soil carbon storage. Sci. Adv. *4*, eaaq1689.

51. Butterbach-Bahl, K., Baggs, E.M., Dannenmann, M., Kiese, R., and Zechmeister-Boltenstern, S. (2013). Nitrous oxide emissions from soils: how well do we understand the processes and their controls? Philos. Trans. R. Soc. B *368*, 20130122.

52. Xia, L., Lam, S., Wolf, B., Kiese, R., Chen, D., and Butterbach-Bahl, K. (2018). Trade-offs between soil carbon sequestration and reactive nitrogen losses under straw return in global agroecosystems. Glob. Change Biol. *12*, 5919–5932.

53. Lindroth, R.L. (2010). Impacts of elevated atmospheric CO_2 and O_3 on forests: phytochemistry, trophic interactions, and ecosystem dynamics. J. Chem. Ecol. *36*, 2–21.

54. Lam, S.K., Chen, D., Norton, R., Armstrong, R., and Mosier, A.R. (2012). Nitrogen dynamics in grain crop and legume pasture systems under

elevated atmospheric carbon dioxide concentration: a meta-analysis. Glob. Change Biol. *18*, 2853–2859.

55. Dijkstra, F., Prior, S., Runion, G., Torbert, H., Tian, H., Lu, C., and Venterea, R. (2012). Effects of elevated carbon dioxide and increased temperature on methane and nitrous oxide fluxes: evidence from field experiments. Front. Ecol. Environ. *10*, 520–527.

56. Greaver, T., Clark, C.M., Compton, J.E., Vallano, D., Talhelm, A., Weaver, C., Band, J., Baron, J., Davidson, E., and Tague, C. (2016). Key ecological responses to nitrogen are altered by climate change. Nat. Clim. Chang. *6*, 836–843.

57. Zhou, L., Zhou, X., Shao, J., Nie, Y., He, Y., Jiang, L., Wu, Z., and Bai, H. (2016). Interactive effects of global change factors on soil respiration and its components: a meta-analysis. Glob. Change Biol. *22*, 3157–3169.

58. van Groenigen, K.J., Osenberg, C.W., Terrer, C., Carrillo, Y., Dijkstra, F.A., Heath, J., Nie, M., Pendall, E., Richard, R.P., and Hungate, B.A. (2017).

Faster turnover of new soil carbon inputs under increased atmospheric CO_2. Glob. Change Biol. *23*, 4420–4429.

59. Viechtbauer, W. (2010). Conducting meta-analyses in R with the metafor package. J. Stat. Softw. *36*, 1–48.

60. Crain, C.M., Kroeker, K., and Halpern, B.S. (2008). Interactive and cumulative effects of multiple human stressors in marine systems. Ecol. Lett. *11*, 1304–1315.

61. Xia, L., Wang, S., and Yan, X. (2014). Effects of long-term straw incorporation on the net global warming potential and the net economic benefit in a rice-wheat cropping system in China. Agric. Ecosyst. Environ. *197*, 118–127.

62. Xia, L., Lam, S.K., Yan, X., and Chen, D. (2017). How does recycling of livestock manure in agroecosystems affect crop productivity, reactive nitrogen losses and soil carbon balance? Environ. Sci. Technol. *51*, 7450–7457.