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Widespread reduction of ozone extremes in storylines of future climate



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High ozone levels harm people and the environment, especially during extreme weather. Climate change is expected to increase the frequency and intensity of these events, exacerbating vegetation-atmosphere interactions. However, current models predict inconsistent responses to warming, potentially due to simplified vegetation representations. We address this uncertainty by incorporating realistic vegetation responses to abiotic stresses into a global atmospheric chemistry model. By constructing storylines of future climate with fixed anthropogenic emissions, we quantify how temperature and humidity changes affect ozone and associated mortality. Here, we show that locally, vegetation and photochemistry often act in concert to amplify ozone pollution extremes, while increased humidity in the free troposphere tends to suppress background ozone levels. The latter effect becomes more dominant with increasing temperatures, leading to a widespread decrease in ozone pollution across the Northern hemisphere. The storyline approach is an effective method for disentangling drivers of air pollution perturbed by climate change.

Tropospheric ozone (O_3) has adverse effects on human health¹ and damages vegetation, reducing ecosystem productivity and crop yields². As a photochemically produced pollutant, O_3 is also a primary driver of the atmosphere's oxidation capacity³. The regional distribution of ground-level O_3 is shaped by precursor emissions, vegetation uptake, and photochemistry, all of which depend on meteorological conditions⁴.

Over continental regions, local O_3 levels typically show a positive correlation with temperature⁵, which is generally well represented in models^{6,7}. However, correlations with humidity vary by location, depending on local chemical environments and land-atmosphere coupling regimes⁸. Additionally, drought stress affects the dry deposition of gases like O_3 on vegetation, which remains challenging for some models to represent accurately^{7,9}. Also, biogenic emissions of O_3 precursors are affected by drought conditions¹⁰ and require incorporation of soil moisture¹¹ or photosynthetic activity¹² for more accurate modelling. Consequently, most Earth system models (ESMs) still lack a consistent representation of land-atmosphere-chemistry interactions, which is key to predict continental ground-level O_3 under climate change¹³.

Assessments of future O_3 pollution usually suggest a climate penalty over populated regions¹⁴. Such studies have relied on probabilistic (risk-based) approaches using ensembles of model simulations.

However, this method is limited by uncertainties in the atmospheric circulation response to climate change¹⁵, such as shifts in the polar jet stream and associated precipitation events. Instead, we use a physical climate event storyline approach to explore how a specific warming level would affect air pollution under the same atmospheric circulation patterns like the ones observed during recent heatwaves. This approach separates thermodynamic effects from circulation changes¹⁶. By nudging the model only toward fields of divergence and vorticity from atmospheric reanalysis data¹⁷, we can reproduce large-scale circulation patterns conducive of recent extreme events and assess Earth system responses under a plausible increased warming^{18,19}. To reduce uncertainty and maintain focus, anthropogenic emissions of O_3 precursors are held constant.

During the summers of 2018, 2019, and 2020, Europe experienced a series of exceptional heat waves. The intensity, persistence, and spatial extent of the 2018 summer heatwave was comparable to the 2003 'mega-heatwave'²⁰. In 2019, Europe recorded its highest ever temperatures, while 2020 was one of the three warmest years on record^{21,22}. In addition, the occurrence of severe droughts in 2018 was not limited to central Europe, but extended to large parts of the Northern Hemisphere (NH)²⁰. The severity of Europe's 2018 heatwave was amplified by soil-moisture feedbacks²³, which

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are known to intensify heatwaves²⁴. Concurrently, vegetation feedbacks in the region exacerbated extreme ozone pollution²⁵.

This study presents, for the first time, physical climate event storylines for air pollution under varying levels of warming, using the global atmospheric chemistry-climate model EMAC. We conduct simulations for 2018–2020 at one *factual* (+1.1K) and two sensitivity warming levels (+2K, +2.75K) relative to pre-industrial conditions. These warming levels represent anomalies in the global mean 2m temperature with respect to pre-industrial values. The simulations separate thermodynamic and dynamic aspects of climate change^{18,19}, with thermodynamic variables (temperature, transpiration, moisture) evolving freely, while dynamic variables (atmospheric vorticity and divergence defining the large-scale circulation) are prescribed. This allows us to isolate the thermodynamic effects of climate change on O₃ extremes, minimising influences from changing weather patterns or stratosphere-troposphere exchange.

Our findings of how ground-level ozone changes in two storylines of future climate are presented in the next section. We then examine the drivers of future O₃ extremes, reassess the O₃-climate penalty, and contrast our results with recent literature. Additionally, we analyse changes in various O₃ pollution metrics across our storylines and assess their impact on human health and terrestrial vegetation. The paper concludes with summarising key findings, including a discussion of the storyline approach and the role of vegetation.

Results

Ozone in warmer climates

The 2018–2020 event storyline of a +2K climate (Fig. 1) show an average 5–10 % decrease of ground-level O₃ over the NH oceans and most NH continents due to changes in background O₃. Over industrial NH regions and throughout the Southern Hemisphere (SH), an O₃ increase of 5–10% occurs, with regional hotspots in India and China where changes can exceed +10%. However, the highest relative O₃ increases occurring in the equatorial Pacific Ocean (globally) are due to the local O₃ minimum in this region, i.e. small baseline values amplify the absolute change (ref. 26, Supplementary Fig. S1).

In the +2.75K world, the O₃ reductions across the NH become more pronounced, with decreases up to 15 % over the oceans, also extending into the equatorial region of the SH. The only exceptions to the widespread O₃ decreases in the NH are found in the most-populated regions of India and China. There, O₃ increases found for the +2K world (Fig. 1a) persist, or only slightly decrease in an even warmer (+2.75K) world (Fig. 1b). On average, these changes exceed the internal model variability of daily O₃ (standardised mean difference: Supplementary Figs. S2, S3, S4) and the results are robust across the 3 years. While the SH O₃ shows significant increases at +2K, these changes are smaller at +2.75K (around 5 %), with positive values occurring

uniformly across the extra-tropics but confined to land areas in the equatorial region.

Globally, the +2.75K warming results in a reduction of the tropospheric ozone burden by about 20 Tg relative to the *factual* climate (371 Tg in 2018, Supplementary Tab. S1). This is in apparent contrast to the analysis of CMIP6 climate projections suggesting an increase in ozone burden due to changes in anthropogenic pollutant emissions and inflow from the lower stratosphere²⁷. These effects are essentially excluded by design in our simulations. The storyline approach we use here allows to disentangle these and other drivers of ozone change in the future. In this framework, we conduct a targeted analysis of the role of vegetation and thermodynamic climate feedbacks for ground-level ozone.

Drivers of ground-level ozone

Ground-level O₃ in the two warmer climates is a result of the (changing) contributions of (1) photochemistry, and (2) dry deposition and (3) entrainment of free tropospheric air in the boundary layer. While the latter depends on the background O₃ levels, the first two processes act locally on O₃ at sub-daily scale. We therefore analyze the daily accumulated values of dry deposition flux and net chemical tendency of ground-level O₃. The spatial distribution of the respective changes remains consistent for the analysed years, underscoring the robustness of the underlying physical and chemical processes.

In the *factual* climate, ground-level O₃ production exceeds the chemical loss in most continental regions. In warmer climates, local drivers push towards increased ground-level O₃ concentrations, particularly in the eastern US and East Asia. For instance, in the +2.75K storyline, boreal summer net chemical production increases by up to 10 ppb/d in these regions (see Fig. 2a). Higher temperatures enhance the radical chemistry, as reflected in the dO₃/dT slope (Fig. 3a), favouring O₃ production. At the same time, elevated HO₂ and OH levels also increase chemical loss, but the net effect remains an overall O₃ increase. The availability of radicals depends on biogenic VOCs and NO_x, which together largely control the levels and cycling of HO₂ and OH. In our storyline approach, it is mainly the variations in plant emissions—driven by drought stress, CO₂-inhibition effects, and temperature changes—that influence the chemical environment (see ‘Methods’). In addition, NO emissions from soil influence the chemical regime, particularly in remote continental environments where they modulate O₃ production efficiency. The notable exception with a strong decrease in O₃ chemical production is over Southern India affected by the summer monsoon. Upon warming, intensified precipitation enhances scavenging of ozone and its precursors²⁸.

Enhanced O₃ levels in central and eastern Europe are linked to increased isoprene emissions from plants (Supplementary Fig. S5). This is consistent with earlier findings⁵. Under high-NO conditions, stronger

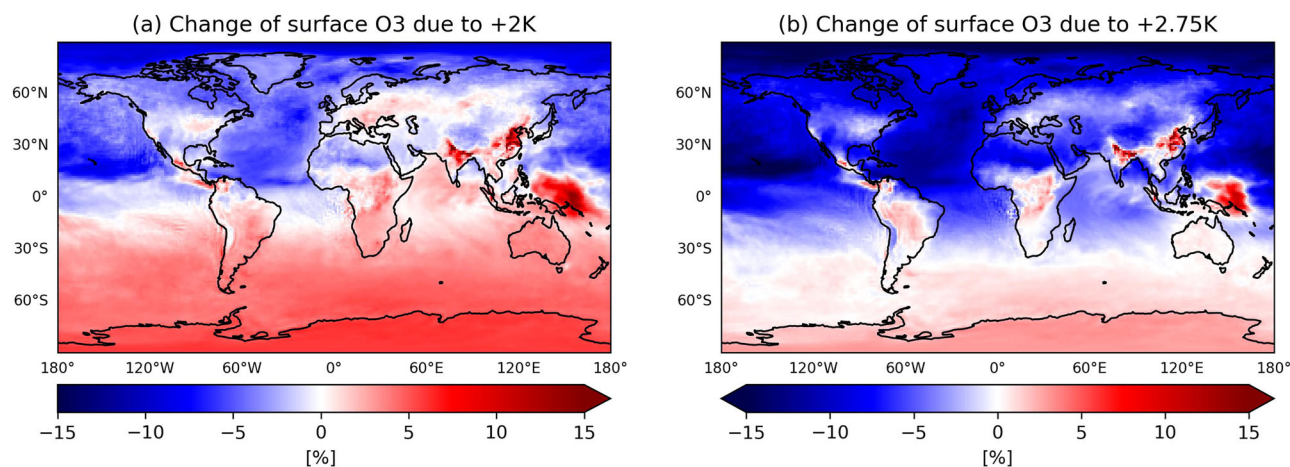


Fig. 1 | Relative difference of daily (24-h) mean ground-level ozone over the 2018–2020 period. Two scenarios (a: +2K vs. *factual*; b: +2.75K vs. *factual*) are shown. Warm and cold colours represent positive and negative changes (NH summer: JJA), respectively.

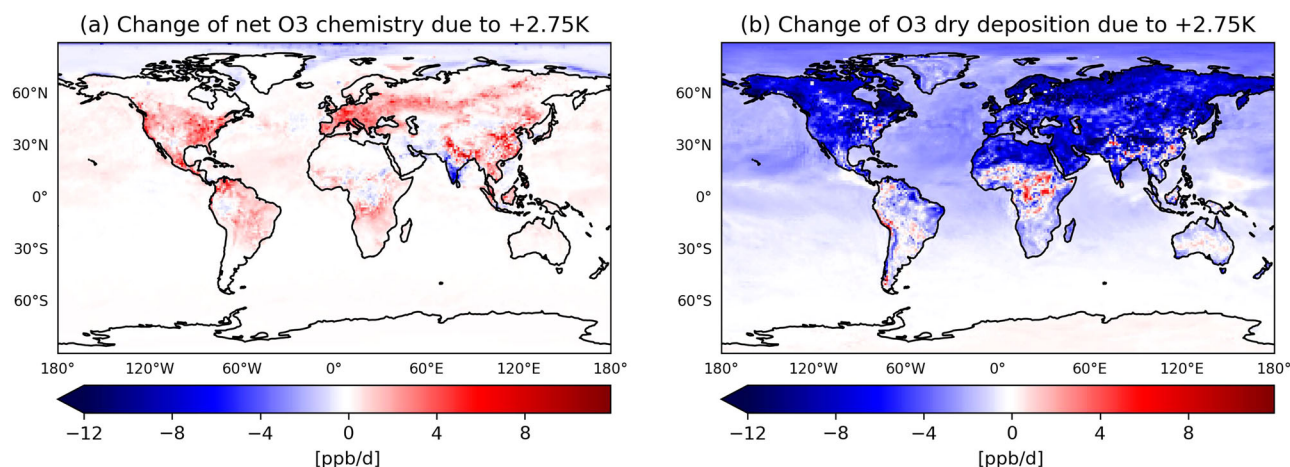


Fig. 2 | Absolute difference of O₃ terms during NH summer (JJA) 2018. a The daily accumulated net O₃ chemistry at ground level and **b** the O₃ dry deposition between the +2.75K and the *factual* climates. Warm and cold colours represent positive and negative changes, respectively.

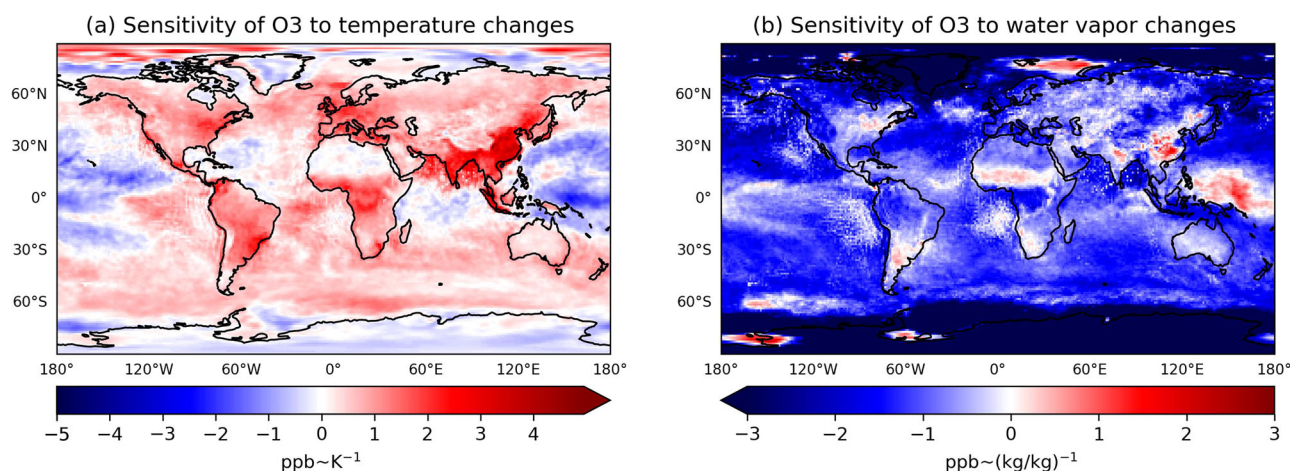


Fig. 3 | Sensitivity of daily mean ground-level O₃ to the warming from the *factual* to the +2.75K climate over the 2018–2020 period. The spatial distribution of linear regression slopes of **a** O₃ vs. temperature changes (dO_3/dT in [ppb/K]) and **b** O₃ vs.

water vapour changes (dO_3/dH_2O [ppb/(kg/kg)]). Warm and cold colours represent the positive and negative O₃ mixing ratio sensitivities, respectively.

isoprene emissions boosts peroxy radical and consequently O₃ formation. However, under low-NO conditions such as above pristine tropical forests, isoprene emissions primarily contribute to O₃ loss, except in regions where increased CO₂ inhibits VOC emissions²⁹.

While daytime O₃ production is enhanced in high-NO_x regions due to increased isoprene-driven radical chemistry, nighttime O₃ loss is also strengthened in a warmer climate due to increased soil NO emissions. During the night, nitrogen oxides (NO) emitted from the soil consume O₃ to form NO₂ and NO₃, eventually producing HNO₃ which is efficiently taken up by vegetation and cannot contribute to daytime ozone production. In the +2.75K world, this stronger nighttime loss reduces the O₃ level relative to the *factual* world, particularly in the SH, where it compensates for up to 20% of the daily O₃ changes (Supplementary Fig. S6). This compensating effect is less pronounced in the NH (other NO sources are more important). Globally, soil NO emissions are calculated to increase by about 0.7 Tg(N)/yr (≈10%) in the future.

While changes in chemical production and loss shape O₃ concentrations in a warmer climate, surface processes such as dry deposition and, specifically, plant uptake also play a crucial role in determining regional O₃ levels. The interactions between vegetation, soil moisture, and atmospheric composition modulate O₃ deposition patterns. In particular, future CO₂ increases reduce plant stomatal opening in regions like the Amazon, Central Africa, and Southeast Asia, leading to reduced

O₃ uptake by vegetation. Conversely, higher humidity enhances the O₃ uptake at leaf surfaces³⁰.

In the +2.75K storyline, O₃ deposition decreases in NH boreal forests. This reduction is primarily explained by increased O₃-induced stress, which causes 20–30% more damage to vegetation, limiting its ability to take up O₃. Additionally, drought stress increases of 5–10%, resulting from higher temperatures⁹, further weakens plant health and increases vulnerability to O₃ exposure. Changes in soil moisture in warmer climates are mostly localised. There is no regional pattern of change in the land-atmosphere coupling regimes and hence in dry deposition velocity. Overall, the change in the actual dry deposition flux (Fig. 2b) is dominated by the changes in the O₃ concentrations (Fig. 1b).

In addition to local drivers, entrainment of background O₃ from the free troposphere is also an important contributor to ground-level O₃. Background ozone is strongly reduced in the +2.75K storyline simulation (Fig. 1b). The changes are most pronounced over the oceans and affect the continents via long-range transport, e.g. from the North Atlantic to Northern Europe³¹. Much of the decrease is due to enhanced O₃ destruction in the free troposphere (Fig. 4), where, in contrast to the (continental) boundary layer, water vapour is the limiting factor for the O(¹D) loss³². This (tropospheric) loss term increases by 11.7% (150 Tg/yr) in the NH and by 6.1% (154 Tg/yr) globally, consistent with findings of widespread decreases in O₃ in warmer and moister climates^{13,33,34}.

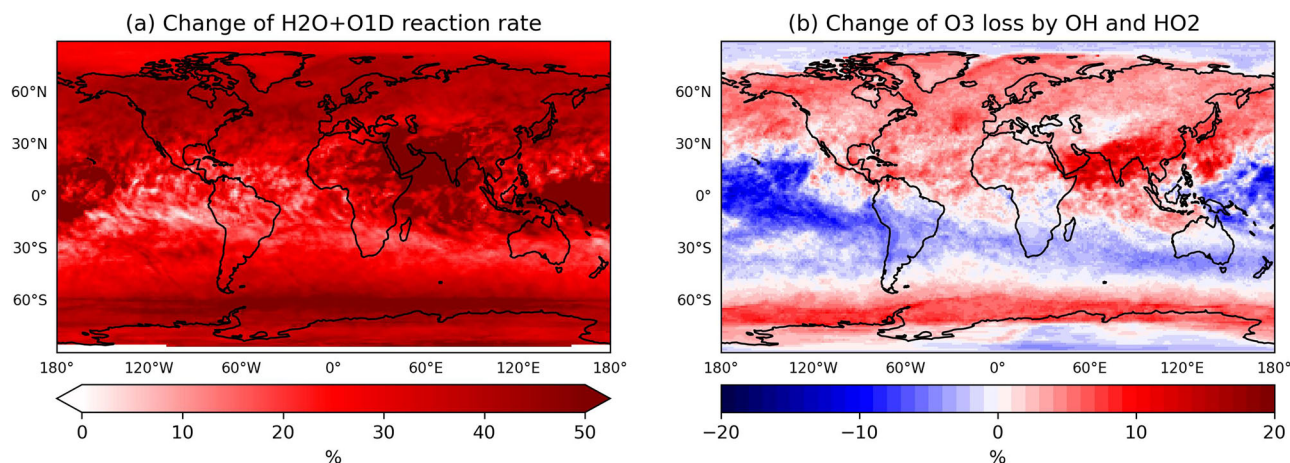


Fig. 4 | Major O₃ losses in the free troposphere. Relative change of the (daily accumulated) **a** $O(^1D)+H_2O$ reaction rate and **b** O₃ loss by OH and $HO_2 \approx 8$ km above the surface due to the +2.75K-climate during summer (JJA) 2018.

The overall changes in ground-level O₃ result from the interplay between local drivers (i.e. photochemistry, precursor emissions, and dry deposition) and reductions in background O₃. The balance of these competing processes varies regionally, highlighting the complexity of ozone-climate interactions.

Ozone-climate penalty or benefit?

The ‘O₃-climate penalty’ refers to the amplification of O₃ air pollution due to global warming, quantified as the rate of O₃ change per unit of temperature change. Several methods for estimating this penalty exist in the literature, such as the long-term correlations between observed O₃ and temperature, or perturbation analyses of temperature changes (ref. 35 and references therein). Here, we estimate this effect using the *factual* EMAC and the +2.75K storyline simulations (Fig. 3).

The sensitivity of O₃ to temperature changes (dO_3/dT) increases significantly, reaching a maximum of +5 ppb/1K in East and South Asia (Fig. 3a). This is consistent with the results by Zanis et al.¹³, which attributes the enhanced sensitivity to stronger anthropogenic NO_x emissions. In contrast to their study, our simulations with fixed anthropogenic emissions indicate that the increase in dO_3/dT at ground level is related to enhanced biogenic VOC emissions in response to warming. Indeed, this relationship has already been observed in China³⁶. The regional increase of O₃ with temperature in the North-East of the US (positive slopes, Fig. 3a), aligns with observations in this region⁷. Over the oceans, we find negative slopes only in the tropical Pacific, in agreement with the CMIP6 model ensemble under the ssp370 scenario¹³.

Future increases of atmospheric water vapour particularly affect the chemistry in the free troposphere³². The sensitivity of O₃ to humidity (dO_3/dH_2O , Fig. 3b) over North Eastern America shows a north-south gradient going from positive to slightly negative values. This is related to a transition of the land-atmosphere coupling from a soil water-limited to a energy-limited regime⁸. Since these coupling regimes do not change for North America in the +2K and +2.75K scenarios, the changes in ground-level O₃ are consistent with the dO_3/dH_2O reported by Kavassalis et al.⁷. A similar reason also applies to East Asia, where positive slopes reflect the shift from a VOC-limited to a NO_x-limited chemical regime³⁷. The concurrent increase of O₃ and H₂O in the tropical West Pacific Ocean is related to strengthening moist convection, which draws humid, O₃-rich air towards regions with low tropospheric O₃ columns³⁸.

While our assessment of future O₃ sensitivities confirms a significant increase in O₃ production with rising temperature—the so-called ‘climate penalty’—here we highlight in addition the importance of the O₃ sensitivity to water vapour. In most regions of the NH with moderate NO_x pollution,

this effect may result in a regional ‘climate benefit’ by counteracting the temperature-driven O₃ increase.

Ozone pollution extremes

We now consider the implications of the simulated O₃ changes for human and ecosystem health in the two storyline experiments. Assessing these impacts requires robust metrics to characterise O₃ extremes, two of which are used here (Fig. 5). While the daily maximum 1-h O₃ mixing ratio (MHM1O₃) captures the short-term variability due to chemical ozone production and loss, governments often refer to the daily maximum 8-h running average of O₃ mixing ratio (MDA8O₃, e.g. the World Health Organisation guidelines³⁹, 50 ppb threshold) as a guideline. MDA8O₃ takes into account the relevant human exposure time to O₃ and is influenced by factors such as dry deposition¹.

Under present-day conditions (equivalent to a +1.1K warming relative to pre-industrial times, the *factual* simulation), the highest number of MHM1O₃ events exceeding 90 ppb falls within the 90–100 ppb range across the studied regions: Europe, South Asia, East U.S. and East Asia (first four histograms, Fig. 5a). East Asia experiences the most extreme events, with a maximum of 1750 occurrences. In most of these regions, the number of MHM1O₃ extremes decreases exponentially as the threshold increases beyond 90 ppb, following a log-normal distribution up to 125 ppb. A significantly higher number of MDA8O₃ than MHM1O₃ events occur in Europe (Fig. 5c), indicating the important role of long-range transport, especially in summer when Asian and North American emissions contribute more to European O₃ levels than regional emissions³¹.

In the +2K storyline, the number of MHM1O₃ events increases across all four regions (Fig. 5a, grey lines). By far the largest increase in number of extreme events (about 800, 13%) is seen in Europe compared to changes of less than 6% in the US and Asia. These regional differences indicate that higher baseline O₃ levels in Europe due to the European heatwaves in summer 2018 is more prompted for increases in O₃ extremes. MDA8O₃ extremes intensify in Europe and Asia, occurring only at high O₃ levels in Europe and at medium levels in South Asia due to the different background pollution and chemistry regimes (Fig. 5c, grey lines).

As the world continues to warm, the O₃ extremes become less frequent in the NH due to a decline in background ozone transported from distant regions. However, in the Tropics and Subtropics this effect is largely offset by an increase in net O₃ production, as most evident in Fig. 5d. In regions like South China, with high-NO and a VOC-limited ozone regime, the strong net O₃ production is fuelled by biogenic VOC emissions especially in summer⁴⁰. Enhanced isoprene emissions as predicted in the two warming scenarios (see Supplementary Fig. S5) further drive O₃ production up that

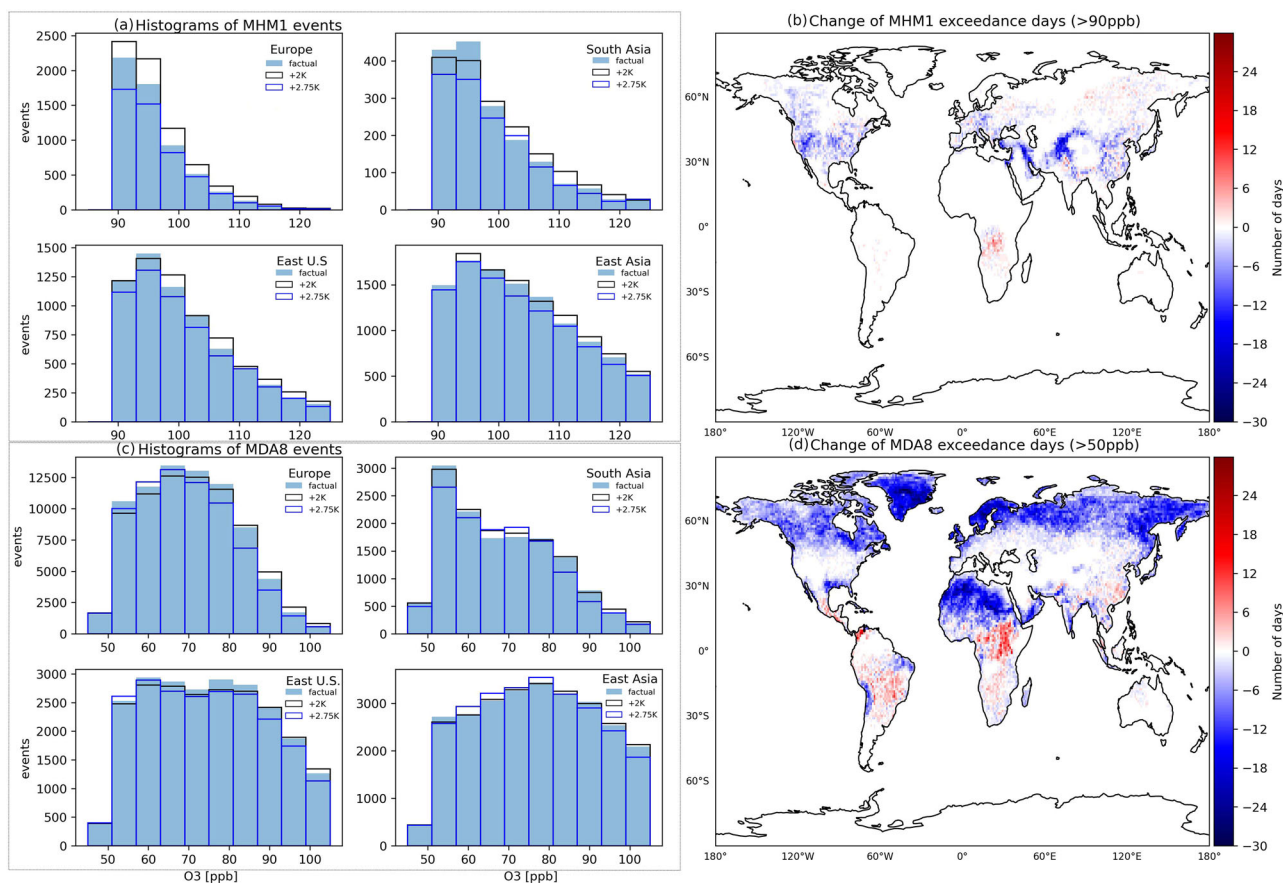


Fig. 5 | O₃ pollution metrics in summer 2018. Number of events for **a** maximum daily 1-h average ground-level O₃ exceeding 90 ppb (MHM1) and **c** daily maximum 8-h running average (MDA8) exceeding 50 ppb in different regions and the corresponding global distribution of the different number of days for **b** MHM1 and **d** MDA8 between +2.75K and *factual* climates. Dark blue solid, black hollow and

blue hollow bars represent *factual*, +2K and +2.75K climates, respectively. The warm and cold colours in the colour bar represent positive and negative changes, respectively. Note that the regions are defined according to the sixth IPCC assessment report⁸⁴: Europe (16,17,18, only land), S.E. U.S.: 5, East Asia: 35, South Asia (SAS): 37.

cannot be completely offset by the entrainment of air with lower background O₃.

Detrimental effects on humans and plants

Finally, we estimate how structural changes in O₃ pollution would affect plants and human health in the +2K and +2.75K storylines, given the same atmospheric circulation as observed in 2018–2020. Note that these impacts are derived for constant anthropogenic pollutant emissions to reduce a key driver of uncertainty in such calculations.

We first assess the cumulative stomatal O₃ uptake using a dynamic threshold (see ‘Methods’), referred to as the phytotoxic ozone damage (POD). Our estimated global distribution, with local values reaching 100 mmol(O₃)/m², is generally in agreement with the estimates by^{41–43}. The highest POD values consistently occur over tropical forests, although they are somewhat overestimated due to a differences between plant types used in our model and those in reality. In the +2.75K storyline, POD significantly increases in NH forests (Fig. 6b), despite an overall decline in summer O₃ levels. This increase in stomatal O₃ fluxes is primarily driven by reduced atmospheric stability, which enhances aerodynamic transport to vegetation and the overall deposition velocity⁴⁴.

We also assess human health impacts by estimating premature mortality due to long-term exposure to elevated O₃ levels (see ‘Methods’). Based on Fig. 7a, the estimated annual total is at 0.14 million O₃-attributable mortality cases, primarily concentrated in China and India, two of the world’s most polluted and densely populated regions. Comparisons with other studies show higher global estimates for 2019 (with 0.42 million deaths

from O₃-attributable chronic respiratory disease and 0.37 million deaths from chronic obstructive pulmonary disease)^{45,46}. These discrepancies likely stem from methodological differences, including the use of a lower relative risk for all-cause mortality in this study (1.01 vs. 1.06 relative risk,⁴⁷) and the use of different O₃ thresholds⁴⁸. Additionally, our estimates may be lower because we assume fixed (‘present-day’) anthropogenic emissions, while other air climate change studies incorporate future emissions scenarios (e.g. ssp3.70: 3.12 million deaths; RCP8.5: 0.32 million deaths)^{49,50}. Relative to the *factual* world, ground-level O₃ in the +2K storyline (Supplementary Fig. S7) leads to a higher mortality worldwide (all-causes, 2018–2020 average). In contrast, in the +2.75K storyline, the corresponding number of deaths decreases in most countries, except for China and several African countries (Fig. 7b), due to a widespread decrease in ground-level O₃ (Fig. 1b). In China, the number of excess deaths increases slightly by 375 across the country, followed by 83 in the Democratic Republic of the Congo, and by 19 in Angola. The differences between the two storylines and the *factual* estimates show that the shift to the warmer climate significantly reduces the global health burden (Supplementary Fig. S7 for +2K and Fig. 7b for +2.75K). India (the world’s most populated country) benefits the most from the shift to the +2.75K storyline, with an estimated 2298 avoided premature deaths (compared to 450 in the USA). Although the number of deaths in China increases under the +2.75K storyline relative to the *factual* climate, the rise is smaller than in the +2K storyline. Overall, the +2.75K storyline prevents 78.2 % of O₃-related deaths globally, highlighting the substantial health benefits of lower ground-level O₃ concentrations in this warmer climate scenario.

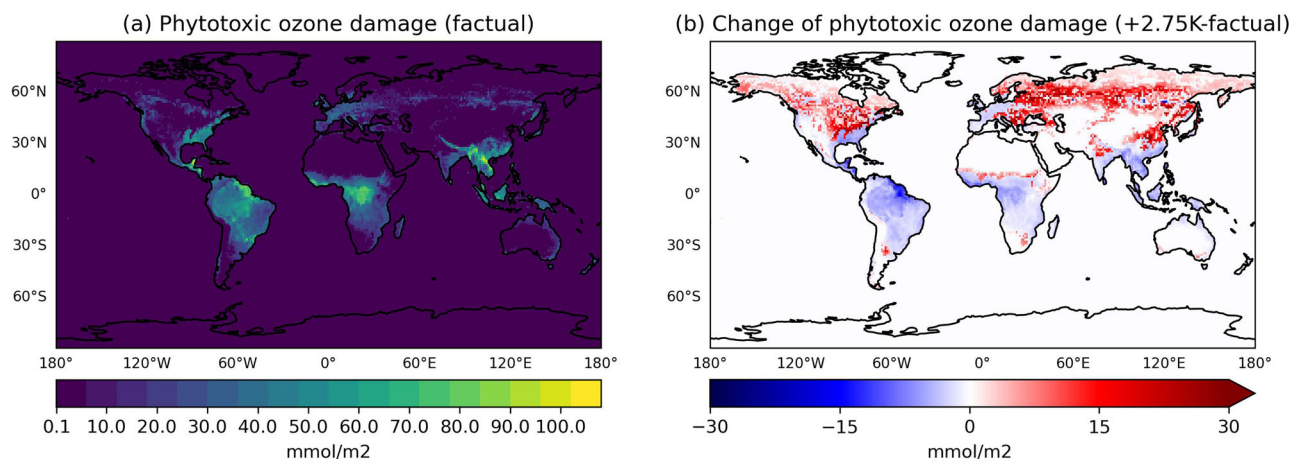


Fig. 6 | Mean phytotoxic ozone damage over the 2018–2020 period. a *factual* climate (+1.1K vs. preindustrial) and **b** absolute difference between the +2.75K climate and *factual* climate. Warm and cold colours in the colour bar represent **a** maximum and minimum values and **b** positive and negative changes, respectively.

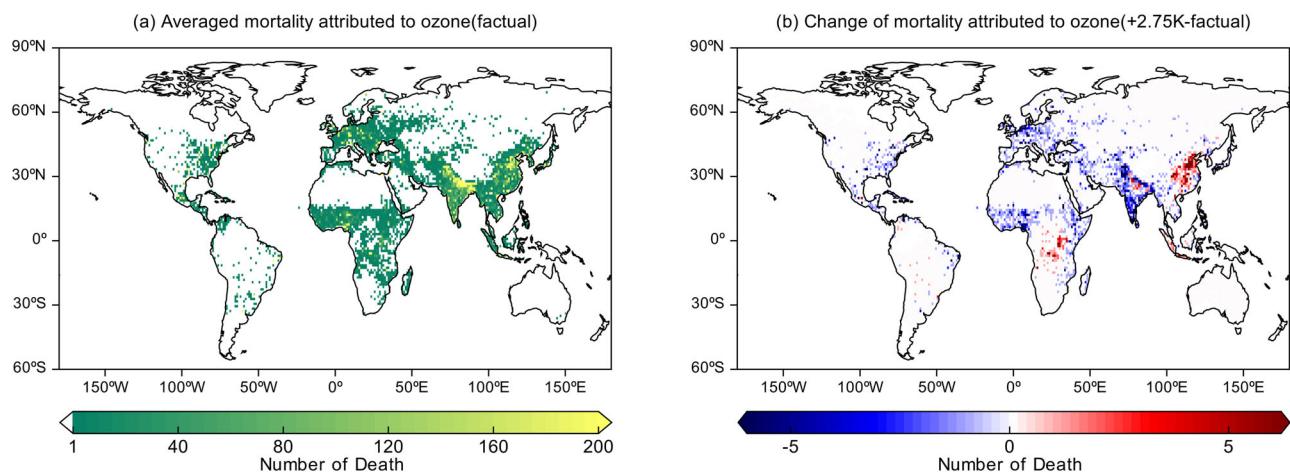


Fig. 7 | Mean premature mortality over the years 2018–2020. The global distribution of a mean premature mortality in *factual* climate (+1.1K vs. preindustrial) and **b** difference of premature mortality between +2.75K and *factual* climates. (Note:

white colour in colour bar (a) means zero, warm and cold colours represent the positive and negative changes, respectively).

Discussion

This study assesses the evolution of ozone pollution in a warmer world by examining two different physical climate event storylines. These storylines are designed to address the question: How would ozone pollution and its impacts under the heatwaves of 2018–2020 have unfolded in a world that is +2K and +2.75K warmer, assuming the same atmospheric circulation patterns? Using fixed anthropogenic emissions, this methodology provides a unique way to focus on the thermodynamic effects of climate change and its impacts on the short-term vegetation response.

Our results show that ozone extremes in the +2K warmer world will increase in many parts (primarily across the SH and in highly populated regions of the NH) due to the amplification of O₃ production by temperature and higher biogenic emissions. In contrast, a warming of +2.75K leads to a dominant increase of the major O₃ loss terms in the NH, resulting in a substantial reduction of O₃ extremes across the NH and the SH equatorial regions, in line with some previous studies (e.g. ref. 13). Thus, the thermodynamic aspects of global warming above +2K indicate (somewhat unexpectedly) potential benefits for air quality, including a reduction of the frequency and intensity of pollution extremes harmful to human and vegetation health. The point of transition from climate penalty to benefit for ground-level ozone over the continents is regionally dependent on many factors like interactions with vegetation and changes in anthropogenic

emissions. Narrowing down the warming levels for these transitions would require many storylines differing for much lower degree of warming. Nevertheless, considering the positive bias global models have⁵¹ and the current air quality policies in Europe and Asia, the onset of ozone-benefit over the continents might happen at a degree of warming much lower than is suggested by +2.75K simulation in this study.

Conducting the first storyline-based air pollution study shows its suitability for extending the knowledge gained from classical probabilistic predictions. A significant uncertainty source in model-aided climate change studies, the uncertainty due to changes in atmospheric dynamics, is largely avoided by the storyline approach, as is the use of fixed anthropogenic emissions. The computational effort, in addition, is significantly reduced compared to conventional probabilistic approaches.

The storyline approach offers a very versatile method for the scientific community to explore future changes in air pollution. By incorporating plant sensitivities to drought and ozone, this study enhances the realism of vegetation-atmosphere interactions, which is crucial for reducing uncertainties in future air quality and climate projections over continents. While our results may be sensitive to parameterisations of the relevant processes (e.g. vegetation response, kinetic chemistry scheme, etc.), our work underscores the need to further study weather-composition interactions in a warming climate. The storyline framework presented here should also be

expanded to account for changes in land cover, wildfire intensity and frequency, and anthropogenic emissions. Disentangling these effects from the changes in atmospheric circulation will ensure a comprehensive understanding of future changes in air pollution.

Methods

Global atmospheric chemistry model

In this study we use the ECHAM/MESSy global atmospheric chemistry model (EMAC), which is based on the fifth-generation European Centre Hamburg general circulation model [ECHAM5, version 5.3.02;⁵²] coupled with the Modular Earth Submodel System (MESSy)^{53,54}. MESSy provides a flexible infrastructure with more than 30 different submodels representing chemical, physical and biogeochemical processes for coupling processes to build comprehensive ESMs. The gas-phase chemistry is based on a comprehensive kinetic mechanism (310 reactions, 155 species) as used in the EMAC simulations for the Chemistry-Climate Model Intercomparison Project (CCMI2)^{55,56}. Due to the complexity of the kinetic chemistry mechanism, the kinetic model is augmented with additional diagnostic of chemical production and loss of compounds of interest⁵⁷. In addition to the conventional odd oxygen family (O_x) approach to estimate atmospheric ozone budget (detailed in Supplement Tab. S1), the production and loss rates of O_3 in different reactions, as well as total production, loss and net rates, are diagnosed in the simulations. The 7-mode aerosol scheme simulates black carbon, organic matter, dust, sea spray, sulphate and ammonium nitrate aerosols⁵⁸. A simplified scheme for secondary organic aerosols production from major precursors is included⁵⁹.

Emissions

The anthropogenic emissions are branched off from the EMAC CCM12 simulation⁵⁶ output for the respective periods. The mixing ratios of long-lived species (greenhouse gases: CO_2 , CH_4 , N_2O etc.) at the lowest model layer are assimilated via Newtonian relaxation (every 3 h) to the projected mixing ratios branched off from the EMAC CCM12 simulation output. The lightning emissions of NO are based on the correlation between the convective cloud top height and the occurrence of flashes and tuned to an average total of 1 Tg(N)/yr in order to achieve realistic present-day O_3 burden in the troposphere. Soil NO emissions are dependent on temperature, precipitation and soil fertilisation and are calculated with the Yiengerle-Levy algorithm⁶⁰. Global emissions in EMAC are about 7 Tg(N)/yr⁶¹. The biogenic emissions of isoprene and other hydrocarbons are modelled with the MEGAN algorithm. The model accounts for the CO_2 inhibition effect using the online calculated CO_2 concentration within the plants⁶². The unstressed emissions fluxes are first calibrated to best estimates by⁶³. Then, a drought stress factor based on the CO_2 assimilation rate is applied according to ref. 12. Final modelled isoprene emissions for the 2018–2020 period are 329 Tg/yr, consistent with⁶⁴.

Abiotic stresses

The transpiration and dry deposition process at canopy scale is represented by a photosynthesis scheme that describes the CO_2 assimilation of plants based on physiological considerations (here only for crops) as a function of CO_2 , temperature, radiation, available and humidity as used in the IFS model⁶⁵. Additionally, two abiotic stressors were implemented for the purpose of this study. First, the plant response to drought stress depends here on leaf water potential which have been shown to succeed over the common used soil-moisture stress factor^{66,67}. The O_3 -induced damage, based on⁶⁸, assesses the O_3 flux derived from the multiple resistance scheme^{69,70} against the phytotoxic threshold. Different from other studies, we implemented a dynamical measure for this based on the gross assimilation of plants and a proportionality constant of $0.20 \mu g(O_3)/mg(CO_2)$ ⁷¹. To consider the limited lifetime of leaves we reset the accumulated ozone damage when the vegetation density decreases consecutively for 2 months. Vegetation density is prescribed with a monthly averages of Leaf Area Index (LAI [m^2/m^2]) obtained by a Moderate Resolution Imaging Spectroradiometer.

Storyline approach

A storyline is a narrative description of a scenario(s) focused on the main characteristics and dynamics, and the relationships between key driving forces (IPCC, <https://www.ipcc-data.org/guidelines/pages/definitions.html>). We follow Shepherd et al.⁷² defining a physical climate event storyline ‘as a physically self-consistent unfolding of past events, or of plausible future events or pathways’. Across the storyline simulations performed here the large-scale dynamics of the recent past (2018–2020) is fixed by nudging only the horizontal winds (via divergence and vorticity) up to an altitude of 12 hPa. Nudging is a Newtonian relaxation of prognostic variables by¹⁷ towards ERA5 meteorological reanalysis data⁷³. It is applied to all wave numbers, up to 106 as the spatial model resolution is T106L47MA ($\approx 1.1^\circ \times 1.1^\circ$). The relaxation times are between 6 and 48 h. These are numbers higher than the ones used by other studies^{18,19} and were chosen to minimise model deviation from observations. Temperature and pressure are not nudged allowing the model to give a thermodynamic response in the warmer climates. The latter are realised by perturbing the forcing associated with:

(1) the ocean for which synthetic sea surface temperatures (SST) patterns consistent with a 2K and 2.75K higher global average temperature are prescribed¹⁸. To this end, we used sea and land surface temperature data simulated by the first ten members of the MPI-ESM ssp245 ensemble (2015–2100) for CMIP6^{74,75}. The 2m temperature anomaly relative to the pre-industrial period 1850–1920 can be found in the supplement (Fig. S12). Following the approach by⁷⁶, the SSTs at year y and month m for the warmer world scenario ($SST_{y,m,+2K}$ and $SST_{y,m,+2.75K}$) are calculated by perturbing the SSTs for the *factual* simulation ($SST_{y,m}^{ERA5}$) with temperature anomalies. The latter are obtained by (a) taking the average SSTs respectively for 2064–2073 ($SST_{+2.75K}^{CMIP6}$) and 2090–2100 ($SST_{+2.75K}^{CMIP6}$) relative to the historic period 1850–1920 ($SST_{y,pi}^{CMIP6}$) and (b) applying a weighting factor w_y , multiplied with the difference of the future and *pi* SST values. This gives the ‘warming pattern’, which is based on normalised near-surface temperatures of the 1850–2100 period. The weighting factor w_y thereby accounts for the fraction of the total warming (relative to a pre-industrial baseline) that has already occurred by year y since pre-industrial time. It considers both the temporal evolution of warming and the spatial variations to reflect regional differences in warming rates. For the +2K scenario the formula is

$$SST_{y,m,+2K} = SST_{y,m}^{ERA5} + w_y(SST_{+2K}^{CMIP6} - SST_{y,pi}^{CMIP6}) \quad (1)$$

(2) The long-lived climate gases (LLCG) CO_2 , CH_4 , N_2O , CFC11, CFC12 are nudged with data branched off from the CCM12-RD2 simulation performed with EMAC⁵⁶. The RD2 CCM12 simulations (several members) reach a global average temperature anomaly of 2.75K at the end of this century, following the SSP2-4.5 CMIP6 scenario (https://www.sparc-climate.org/wp-content/uploads/sites/5/2021/07/SPARCnewsletter_Jul2021_web.pdf). Global average time series of LLCG are shown in Supplementary Fig. S13. Anthropogenic emissions of short-lived gases are fixed to the year 2018. The land cover classification is fixed and the leaf area index is prescribed with MODIS data for 2018 (and following years)⁷⁷.

Ozone bias in the *factual* simulation

The comparison of tropospheric ozone with the chemical reanalysis TES⁷⁸ in Supplementary Fig. S8 (mean of data in 700–800 hPa) shows a mean bias of 10–20 ppb, exemplary for 2018, which is in agreement with the literature. The overestimation over the Indian ocean is due to complex cloud chemistry neglected here²⁸. Additionally, high biases occur over mountain ranges (Andes, California, Mongolia) which is linked to the limited model resolution of topography. However, this is a long-standing issue for global models. A comparison of 6 CMIP6 models with ground-level O_3 observations (TOAR) show a consistent overestimation of 16 ppb in most regions (NH) during the years 2005–2014⁷⁹ similar to⁵¹. The extremes for ground-level O_3 simulated by EMAC are compared to TOAR data gridded at $1^\circ \times 1^\circ$

spatial resolution. The daily 1 h-max values for 2018 are shown for Europe, China and US (Supplementary Figs. S9, S10, S11). EMAC generally overestimate also the continental ozone extremes in the NH. The biases are larger (up to 10 ppb) in boreal summer. Over China the biases are the smallest. The global burden of 370 Tg estimated by EMAC for the *factual* (+1.1K vs. pre-industrial) climate is within the multi-model estimates (330–380 Tg) from the literature^{27,51}. In comparison, EMAC yields about slightly larger chemical production and loss of ozone (see Supplementary Tab. S1). Furthermore, EMAC estimates a lower dry deposition term of 780 Tg/yr (compared to 800–1000 Tg/yr,^{27,51}) due to the additional environmental stressors.

Assessment of premature mortality

We determined the premature mortality number associated with ground-level ozone by applying health impact functions that link variations in air pollution levels to shifts in mortality. The global ground-level ozone concentrations we used are simulated in the EMAC model. Health impact functions for ozone are constructed based on a logarithmic-linear relationship between relative risk (RR) and concentrations, which is defined and widely used in epidemiological research^{80,81}:

$$RR = \exp[\beta(C - C_0)], C > C_0 \quad (2)$$

where β is the concentration-response parameter indicating the additional all non-accidental mortality attributed per unit increase of air pollutant when it is above the threshold concentration. Here, the β value for long-term ozone exposure is 1.0% per 10 $\mu\text{g}/\text{m}^3$ in the peak-season average of daily maximum 8-h mean ozone concentration, which is recommended by Huangfu et al.⁴⁷. C is the simulated ground-level ozone concentration and C_0 is the recommended value by the World Health Organization (WHO) air quality guidelines³⁹ for long-term exposure to ozone. The attributable fraction (AF), which represents the share of the mortality burden associated with the risk factor, was defined as follows:

$$AF = (RR - 1)/RR \quad (3)$$

$$\Delta M = AF \times P \times BMR \quad (4)$$

AF yields an estimate for the additional deaths (ΔM) when multiplied by the baseline mortality rate (BMR, downloaded from <https://data.worldbank.org/indicator/SP.DYN.CDRT.IN>) and the exposed population size (P , accessed from <https://hub.worldpop.org/project/categories?id=3>).

Data availability

The data produced and analysed for this study are available upon request. The Modular Earth Submodel System (MESSy) is continuously further developed and applied by a consortium of institutions. The usage of MESSy and access to the source code is licensed to all affiliates of institutions which are members of the MESSy Consortium. Institutions can become a member of the MESSy Consortium by signing the MESSy Memorandum of Understanding. More information can be found on the MESSy Consortium Website (www.messy-interface.org).

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Author contributions

D.T. and T.E. conceptualised and planned the study. D.T. and S.G. developed the approach for chemical budgeting of ozone. T.E. implemented several model developments, prepared and conducted the global simulations, analysed the model output and wrote most of the manuscript. F.S. estimated the premature mortality related to ozone and the changes in a warmer climate and wrote the respective part of the manuscript, under guidance from M.I.H. F.S. and D.T. evaluated the model performance against surface ozone observations. All authors reviewed and edited the manuscript.

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Competing interests

The authors declare no competing interests.

Additional information

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