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Ozone and haze pollution weakens net primary productivity in China

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Abstract

Atmospheric pollutants have both beneficial and detrimental effects on carbon uptake by land ecosystems. Surface ozone damages leaf photosynthesis by oxidizing plant cells, while aerosols promote carbon uptake by increasing diffuse radiation and exert additional influences through concomitant perturbations to meteorology and hydrology. China is currently the world's largest emitter of both carbon dioxide and short-lived air pollutants. The land ecosystems of China are estimated to provide a carbon sink, but it remains unclear whether air pollution acts to inhibit or promote carbon uptake. Here, we employ Earth system modeling and multiple measurement datasets to assess the separate and combined effects of anthropogenic ozone and aerosol pollution on net primary productivity (NPP) in China. In the present day, air pollution reduces annual NPP by 0.4 Pg C (9%), resulting from a decrease of 0.6 Pg C (14%) by ozone damage, and an increase of 0.2 Pg C (5%) by aerosol direct effects. The enhancement by aerosols is a combination of diffuse radiation fertilization, reduced canopy temperatures, and reduced evaporation leading to higher soil moisture. However, precipitation inhibition from combined aerosol direct and indirect effects reduces annual NPP by 0.2 Pg C (4%), leading to a net air pollution suppression of 0.8 Pg C (16%). Our results reveal strong dampening effects of air pollution on the land carbon uptake in China today. Following the current legislation emission scenario, this suppression will not alleviate by the year 2030, mainly due to a continuing increase in surface ozone. However, the maximum technically feasible reduction scenario could drastically relieve the current level of NPP damage by 70% in 2030, offering protection of this critical ecosystem service and the mitigation of long-term global warming.

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1 Introduction

Surface ozone (O_3) and atmospheric aerosols influence land ecosystem carbon uptake both directly and indirectly through Earth system interactions. O_3 reduces plant photosynthesis directly through stomatal uptake. The level of damage is dependent on both surface ozone concentrations ($[O_3]$) and the stomatal conductance (g_s), the latter of which is closely related to the photosynthetic rate (Reich and Amundson, 1985; Sitch et al., 2007; Ainsworth et al., 2012). The impact of aerosol pollution on vegetation is less clear. Atmospheric aerosols influence plant photosynthesis through perturbations to radiation, meteorology, and cloud. Observations (Cirino et al., 2014; Strada et al., 2015) suggest that an increase in diffuse light partitioning in response to a moderate aerosol loading can improve canopy light use efficiency (LUE) and promote photosynthesis, known as diffuse radiation fertilization (DRF), as long as the total light availability is not compromised (Kanniah et al., 2012). Atmospheric aerosols also reduce leaf temperature (Steiner and Chameides, 2005; Cirino et al., 2014), but the consequence for photosynthesis depends on the relationship between the local environmental temperature and the photosynthetic optimum temperature of approximately 25°C. Aerosol-induced changes in evaporation and precipitation are interconnected but impose opposite effects on photosynthesis; less evaporation preserves soil moisture in the short term but may decrease local rainfall (Spracklen et al., 2012) and lead to drought conditions in the long term. Furthermore, aerosol indirect effects (AIE) on cloud properties can either exacerbate or alleviate the above feedbacks.

China is currently the world's largest emitter of both carbon dioxide and short-lived air pollutants (http://gains.iiasa.ac.at/models/). The land ecosystems of China are estimated to provide a carbon sink (Piao et al., 2009), but it remains unclear how air pollution may affect this sink through the atmospheric influences on regional carbon uptake. O_3 damages to photosynthesis, including those in China, have been quantified in hundreds of measurements (Table S1), but are limited to individual plant species and specific $[O_3]$. Previous regional modeling of O_3 vegetation damage (e.g., Ren et al., 2011; Tian et al., 2011) does not always take advantage of valuable observations and typically the derived

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results have not been properly validated. The aerosol effects on photosynthesis are less understood. Most of the limited observation-based studies (Rocha et al., 2004; Cirino et al., 2014; Strada et al., 2015) rely on long-term flux measurements or satellite retrievals, which are unable to unravel impacts of changes in the associated meteorological and hydrological forcings. Modeling studies focus mainly on the aerosol-induced enhancement in diffuse radiation (e.g., Cohan et al., 2002; Gu et al., 2003; Mercado et al., 2009), but ignore other direct and indirect feedbacks such as changes in temperature and precipitation. Finally, no studies have investigated the combined effects of O_3 and aerosols or how the air pollution influences may vary in response to future emission regulations and climate change.

In this study, we assess the impacts of O_3 and aerosols on land carbon uptake in China using the global Earth system model NASA GISS ModelE2 that embeds the Yale Interactive Terrestrial Biosphere model (YIBs). This framework is known as NASA ModelE2-YIBs and fully couples the land carbon-oxidant-aerosol-climate system (Schmidt et al., 2014; Yue and Unger, 2015). The global-scale model accounts for long-range transport of pollution and large-scale feedbacks in physical climate change. The coupled Earth system simulations apply present-day and future pollution emission inventories from the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS) integrated assessment model (http://gains.iiasa.ac.at/models/). The simulations include process-based, mechanistic photosynthetic responses to physical climate change, O_3 stomatal uptake, carbon dioxide (CO_2) fertilization, and aerosol radiative perturbations, but not aerosol and acid deposition (Table 1). The O_3 and aerosol haze effects on the land carbon cycle fluxes occur predominantly through changes to gross primary productivity (GPP) and net primary productivity (NPP). Therefore, the current study focuses on GPP and NPP impacts and does not address changes in net ecosystem exchange (NEE).

2 Methods

2.1 YIBs vegetation model

The YIBs model applies the well-established Farquhar and Ball-Berry models (Farquhar et al., 1980; Ball et al., 1987) to calculate leaf photosynthesis and stomatal conductance, and adopts a canopy radiation scheme (Spitters, 1986) to separate diffuse and direct light for sunlit and shaded leaves. The assimilated carbon is dynamically allocated and stored to support leaf development (changes in leaf area index, LAI) and tree growth (changes in height). A process-based soil respiration scheme that considers carbon flows among 12 biogeochemical pools is included to simulate carbon exchange for the whole ecosystem (Yue and Unger, 2015). Similar to many terrestrial models (Schaefer et al., 2012), the current version of YIBs does not include a dynamic N cycle. Except for this deficit, the vegetation model can reasonably simulate ecosystem responses to changes in [CO_2], meteorology, phenology, and land cover (Yue et al., 2015). A semi-mechanistic O_3 vegetation damage scheme (Sitch et al., 2007) is implemented to quantify responses of photosynthesis and stomatal conductance to O_3 (Yue and Unger, 2014).

The YIBs model can be used in three different configurations: site-level, global/regional offline, and online within ModelE2-YIBs (Yue and Unger, 2015). The offline version is driven with hourly 1°×1° meteorological forcings from either the NASA Modern Era Retrospective-analysis for Research and Applications (MERRA) (Rienecker et al., 2011) or the interpolated output from ModelE2-YIBs. The online YIBs model is coupled with the climate model NASA ModelE2 (Schmidt et al., 2014), which considers the interplay among meteorology, radiation, atmospheric chemistry, and plant photosynthesis at each time step. For both global and regional simulations, 8 plant functional types (PFTs) are considered (Fig. S1). This land cover data set is derived based on retrievals from both the Moderate Resolution Imaging Spectroradiometer (MODIS) (Hansen et al., 2003) and the Advanced Very High Resolution Radiometer (AVHRR) (Defries et al., 2000). The same vegetation cover is used by the Community Land Model (CLM) (Oleson et al., 2010).

Both the online and offline YIBs models have been extensively evaluated with site-level measurements from 145 globally-dispersed flux tower sites, long-term gridded benchmark products, and multiple satellite retrievals of LAI, tree height, phenology, and

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carbon fluxes (Yue and Unger, 2015; Yue et al., 2015). Driven with meteorological reanalyses, the offline YIBs vegetation model estimates a global GPP of 122.3 ± 3.1 Pg C yr^{-1}, NPP of 63.6 ± 1.9 Pg C yr^{-1}, and NEE of -2.4 ± 0.7 Pg C yr^{-1} for 1980-2011, consistent with an ensemble of land models (Yue and Unger, 2015). The online simulations with ModelE2-YIBs, including both aerosol effects and O_3 damage, yield a global GPP of 125.8 ± 3.1 Pg C yr^{-1}, NPP of 63.2 ± 0.4 Pg C yr^{-1}, and NEE of -3.0 ± 0.4 Pg C yr^{-1} under present day conditions.

2.2 NASA ModelE2-YIBs model

The NASA ModelE2-YIBs is a fully coupled chemistry-carbon-climate model with horizontal resolution of 2°×2.5° latitude by longitude and 40 vertical levels extending to 0.1 hPa. The model simulates gas-phase chemistry (NO_x, HO_x, O_x, CO, CH_4, NMVOCs), aerosols (sulfate, nitrate, elemental and organic carbon, dust, sea salt), and their interactions (Schmidt et al., 2014). Modeled oxidants influence the photochemical formation of secondary aerosol species (sulfate, nitrate, secondary organic aerosol). In turn, modeled aerosols affect photolysis rates in the online gas-phase chemistry (Schmidt et al., 2014). Heterogeneous chemistry on dust surfaces is represented (Bauer et al., 2007). The embedded radiation package includes both direct and indirect (Menon and Rotstayn, 2006) radiative effects of aerosols and considers absorption by multiple GHGs. Simulated surface solar radiation exhibits the lowest model-to-observation biases compared with the other 20 IPCC-class climate models (Wild et al., 2013). Simulated meteorological and hydrological variables have been full validated against observations and reanalysis products (Schmidt et al., 2014).

2.3 Emissions

We use global annual anthropogenic pollution inventories from the GAINS integrated assessment model (Amann et al., 2011), which compiles historic emissions of air pollutants for each country based on available international emission inventories and national information from individual countries. Inter-comparison (Fig. S2) shows that the

GAINS V4a inventory has similar emission intensity (within ±10%) in China as IPCC RCP8.5 scenario (van Vuuren et al., 2011) for most species, except for ammonia, which is higher by 80% in GAINS. The GAINS inventory also projects medium-term variations of future emissions at five-year intervals to the year 2030. The current legislation emissions (CLE) scenario applies full implementation of national legislation affecting air pollution emissions; for China, this represents the 11th five-year plan, including known failures. By 2030, in the CLE inventory, CO decreases by 18%, SO_2 by 21%, black carbon by 28%, and organic carbon by 41%, but NO_x increases by 20%, ammonia by 22%, and non-methane volatile organic compounds (NMVOC) by 6%, relative to the 2010 emission magnitude in China. To account for potential rapid changes in policy and legislation, we apply the maximum technically feasible reduction (MTFR) emission scenario as the lower limit of future air pollution. The MTFR scenario implements all currently available control technologies, disregarding implementation barriers and costs. With this scenario, the 2030 emissions of NO_x decrease by 76%, CO by 79%, SO_2 by 67%, black carbon by 81%, organic carbon by 89%, ammonia by 65%, and NMVOC by 62% in China, indicating large improvement of air quality. Biomass burning emissions, adopted from the IPCC RCP8.5 scenario (van Vuuren et al., 2011), decrease by 1-2% for all pollution species at the year 2030 compared with 2010.

Natural emissions of air pollution show much smaller changes by 2030 compared with anthropogenic emissions. Lightning NO_x emissions increase by ~4% (0.02 Tg N) and soil NO_x emissions increase by ~1% (0.001 Tg N). Emissions of biogenic VOCs, predicted with a photosynthesis-based scheme (Yue et al., 2015), increase by 5% (+0.39 Tg C) for isoprene but decrease by 5% (-0.25 Tg C) for monoterpenes. Changes in natural emissions of SO_2 (volcano and ocean sources) and NH_3 (ocean source) are trivial over China.

2.4 Simulations

2.4.1 NASA ModelE2-YIBs online

We perform 24 time-slice simulations to explore the interactive impacts of O_3 and aerosols on land carbon uptake (Table S2). All simulations are performed in atmosphere-only configuration. In these experiments, [O_3] and aerosol loading are dynamically predicted, and atmospheric chemistry processes are fully two-way coupled to the meteorology and the land biosphere. Simulations can be divided into two groups, depending on whether AIE are included. In each group, three subgroups are defined with the emission inventories of GAINS 2010, CLE 2030, and MTFR 2030 scenarios. In each subgroup, one baseline experiment is set up with only natural emissions. The other three implement all natural and anthropogenic sources of emissions, but apply different levels of O_3 damage including none, low sensitivity, and high sensitivity. To compare the differences between online and offline O_3 damage, we perform two additional simulations, G10ALLHO3_OFF and G10ALLLO3_OFF, which do not account for the feedbacks of O_3-induced changes in biometeorology, plant growth, and ecosystem physiology. We use prescribed sea surface temperature (SST) and sea ice distributions simulated by ModelE2 under the IPCC RCP8.5 scenario (van Vuuren et al., 2011). For these boundary conditions, we apply the monthly-varying decadal average of 2006-2015 for 2010 simulations and that of 2026-2035 for 2030 simulations. Well-mixed GHG concentrations are also adopted from the RCP8.5 scenario. Land cover change projections for this scenario suggest only minor changes between the years 2010 and 2030; for example, the expansion of 3% for grassland is offset by the losses of 1% for cropland and 4% for tropical rainforest. As a result, we elect to apply the same land cover, which is derived from satellite retrievals, for both present-day and future simulations (Fig. S1). We use present-day equilibrium tree height derived from a 30-year spinup procedure (Yue and Unger, 2015) as the initial condition. All simulations are performed for 20 years, and the last 15 years are used for analyses. For simulations including effects of CO_2 fertilization, climate change, and O_3 damages, GPP and NPP reach new equilibrium within 5 years while those for NEE may require several decades due to the slow responses of the soil carbon pools (Fig. S3).

2.4.2 YIBs offline with MERRA meteorology

We perform 15 simulations to evaluate the skill of the O_3 damage scheme for vegetation in China (Table S3). Each run is driven with hourly meteorological forcings from NASA GMAO MERRA (Rienecker et al., 2011). One baseline simulation is performed without inclusion of any O_3 damage. The others, seven runs in each of two groups, are driven with fixed [O_3] at 20, 40, 60, 80, 100, 120, and 140 ppbv, respectively, using either low or high O_3 sensitivities defined by (Sitch et al., 2007). Thus, [O_3] in these offline runs is fixed without seasonal and diurnal variations to mimic field experiments that usually apply a constant level of [O_3] during the test period. We compare the O_3-affected GPP with the O_3-free GPP from the baseline simulation to derive the damaging percentages to GPP, which are compared with values for different PFTs from an ensemble of published literature results (Table S1). All simulations are performed for 1995-2011, and the last 10 years are used for analyses.

2.4.3 YIBs offline with ModelE2-YIBs meteorology

Using the offline YIBs vegetation model driven with ModelE2-YIBs meteorology, we perform 30 simulations to isolate the impacts of aerosol-induced changes in the individual meteorological drivers on carbon uptake (Table S4). Experiments are categorized into two groups, depending on whether the GCM forcings include AIE or not. In each group, three subgroups of simulations are set up with different meteorology for GAINS 2010, CLE 2030, and MTFR 2030 scenarios. Within each subgroup, five runs are designed with the different combinations of GCM forcings. One baseline run is forced with meteorology simulated without anthropogenic aerosols. The other four are additionally driven with aerosol-induced perturbations in temperature, PAR, soil moisture, or the combination of the above three variables. The differences of NPP between sensitivity and baseline runs represent contributions of individual or total aerosol effects. Each simulation is performed for 10 years, with the last 3 years used for analyses.

3 Results

3.1 Evaluation of ModelE-YIBs over China

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3.1.1 Land carbon fluxes: GPP and NPP

To validate simulated GPP, we use a gridded benchmark product for 2009-2011 upscaled from *in situ* FLUXNET measurements (Jung et al., 2009). For NPP, we use a MODIS satellite-derived product for 2009-2011 (Zhao et al., 2005). Both datasets are interpolated to the same resolution of 2°×2.5° as ModelE2-YIBs. Simulated GPP and NPP reproduce the observed spatial patterns with high correlation coefficients (R=0.46-0.86, $p < 0.001$) and low model-to-observation biases (< 20%) (Fig. 1 and Fig. S4). High values of the land carbon fluxes are predicted in the East and the Northeast, where forests and croplands dominate (Fig. S1).

3.1.2 Surface air pollution and AOD

For surface concentrations of $PM_{2.5}$ and O_3, we use ground measurements available for 2014 from 188 sites operated by the Ministry of Environmental Protection of China (http://www.aqicn.org/). In addition, we use rural [O_3] from published literature (Table S5) to evaluate the model. For AOD, we use gridded observations of 2008-2012 from MODIS retrievals. The model simulates reasonable magnitude and spatial distribution of AOD and surface $PM_{2.5}$ concentrations (Fig. 2). Long-term measurements of [O_3] are very limited in China. Comparisons with the 2014 one-year data from 188 urban sites show that simulated [O_3] reproduces reasonable spatial distribution but overestimates the average concentration by >40%. Such discrepancy is in part attributed to the sampling biases, because urban [O_3] is likely lower than rural [O_3] due to high NO_x emissions (NO_x titration) and aerosol loading (light extinction) in cities. Evaluations at rural sites (Table S5) better match the observations that represent the major domain of China (Fig. 3).

3.1.3 Shortwave radiation

We use ground-based observations of surface shortwave radiation and diffuse fraction from 106 pyranometer sites managed by the Climate Data Center, Chinese Meteorological Administration (Xia, 2010). Site selection is based on the availability of continuous monthly measurements during 2008-2012, resulting in 95 sites for the evaluation of total shortwave radiation. For diffuse radiation, we select the 17 sites only that provide continuous measurements during 2008-2012. Simulated surface shortwave

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radiation and diffuse fraction agree well with measurements at 106 sites, especially over the East where low solar insolation while high diffusion are predicted (Fig. 4).

3.1.4 Ozone vegetation damage function

We adopt the same approach as Yue et al. (2016) by comparing simulated GPP-to-[O_3] responses (Table S3) with observations from multiple published literature (Table S1). We aggregate these measurements into six categories, including evergreen needleleaf forest (ENF), deciduous broadleaf forest (DBF), shrubland, C3 herbs, C4 herbs, and a mixture of all above species. We derive the sensitivity of GPP to varied [O_3] (Fig. 5) using the YIBs offline version. For most PFTs, simulated O_3 damage increases with [O_3] in broad agreement with measurements. Predicted O_3 damage reproduces observations with a correlation coefficient of 0.61 (for all samplings, n=32) and in similar magnitudes (-17.7% vs. -20.4%), suggesting that the damage scheme we adopted from Sitch et al. (2007) is ready to use in China. For the same level of [O_3], deciduous trees suffer larger damages than evergreen trees because the former species are usually more sensitive (Sitch et al., 2007) and have higher g_s (therefore higher uptake) (Wittig et al., 2007). Flux-based O_3 sensitivity for C4 herbs is only half that of C3 herbs (Sitch et al., 2007), however, concentration-based O_3 damages, both observed and simulated, are larger for C4 plants because of their higher uptake efficiency following high g_s (Yue and Unger, 2014).

3.2 O_3 effects in China

We focus our study domain in eastern China (21°-38°N, 102°-122°E, including the North China Plain, the Yangtze River Delta, and part of the Sichuan Basin), a region that suffers from high levels of O_3 and aerosols from anthropogenic pollution sources (>75% contribution; Fig. S5). We estimate that surface O_3 decreases annual GPP in China by 10.3% based on YIBs offline simulations in the absence of feedbacks from O_3 vegetation damage to meteorology and plant growth. The damage is stronger in summer, when the average GPP decreases by ~20% for both deciduous trees and C3 herbs in the East (Fig. 6). In contrast, a lower average damage to GPP of ~10% is predicted for evergreen

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needleleaf trees (because of low sensitivity) and C4 herbs (because of the mismatched spatial locations between C4 plants and high [O_3], Fig. S1 and Fig. 2d).

O_3 damage to photosynthesis can influence plant growth. At the same time, the O_3-induced reductions in stomatal conductance can increase canopy temperature and inhibit plant transpiration (Lombardozzi et al., 2015), leading to surface warming and rainfall deficit (Fig. S6). These biometeorological feedbacks may in turn exacerbate the dampening of land carbon uptake. Application of ModelE2-YIBs that allows for these feedbacks gives an O_3-induced damage to annual GPP of 10.7%, a similar level to the damage computed in YIBs offline. Sensitivity simulations (offline and online) with zero anthropogenic emissions show no O_3 damage, because the [O_3] exposure from natural sources alone is lower than the damaging threshold of 40 ppbv for most PFTs (Fig. 5). Our results indicate that present-day surface O_3 inhibition, averaged for low and high plant sensitivities, reduces total NPP in China by 0.59 ± 0.11 Pg C yr^{-1} (Table 2), assuming no direct impacts of O_3 on plant respiration.

3.3 Aerosol haze effects in China

Aerosols decrease direct solar radiation but increase diffuse radiation (Fig. S7), the latter of which is beneficial for canopy photosynthesis. The online coupled model quantifies the concomitant meteorological and hydrological feedbacks (Fig. 7) that further influence the radiative and land carbon fluxes. Reduced insolation decreases summer surface temperature by 0.63°C in the East, inhibiting evaporation but increasing relative humidity (RH) due to lower saturation vapor pressure (Table S6). These feedbacks combine to stimulate photosynthesis (Fig. 8a), which, in turn, strengthens plant transpiration and further increases RH, contributing to enhanced precipitation and cloud cover. Moreover, soil moisture increases with lower evaporation and higher precipitation. Inclusion of AIE results in distinct climatic feedbacks (Fig. S8). The AIE reduces cloud droplet size and inhibits summer precipitation by 0.9 mm day^{-1} (13%), leading to a 3% decline in soil moisture (Table S6). The AIE also lengthens cloud lifetime and increases cloud cover, further reducing available radiation and causing a stronger surface cooling.

We separate the relative impacts due to aerosol-induced perturbations in temperature, radiation, and soil moisture (Fig. 8). The overall impact of the aerosol-induced biometeorological feedbacks on the carbon uptake depends on the season and vegetation type. In the summer, the aerosol-induced surface cooling brings leaf temperature closer to the photosynthetic optimum of 25°C, DRF enhances light availability of shaded leaves and LUE of sunlit leaves, and the wetter soil alleviates water stress for stoma. Consequently, aerosol-induced hydroclimatic feedbacks promote ecosystem NPP, albeit with substantial spatiotemporal variability (Fig. 8a and Table 2). Surface cooling enhances NPP in summer (Fig. 8b) but induces neutral net impacts on NPP in spring and autumn (not shown), when leaf temperature is usually below 25°C, because the cooling-driven reductions of photosynthesis are accompanied by simultaneous reductions in plant respiration. Plant phenology is responding to aerosol-induced cooling; however, such changes alone exert much smaller impacts on the ecosystem carbon uptake compared to those driven directly by temperature changes (Yue et al., 2015). We find strong aerosol DRF in the Southeast and the Northeast, where AOD is moderate (Fig. 8c). Over the North China Plain and the Southwest, aerosol DRF is more limited. In these regions, the local background aerosol layer and/or cloud over are sufficiently optically thick that the effect of anthropogenic aerosol pollution is largely to attenuate direct sunlight and reduce NPP (Cohan et al., 2002). Aerosol-induced cooling increases soil moisture over most of the East (Fig. 7f), but the beneficial responses are confined to the Central East (Fig. 8d), where C3 crops dominate (Fig. S1). These short-root plants are more sensitive to short-term water availability than deep-root trees (Beer et al., 2010; Yue et al., 2015).

In contrast, inclusion of AIE results in detrimental impacts on NPP (Table 2). Aerosol-induced drought strongly reduces regional NPP especially over the Northeast and North China Plain (Fig. S9d), where cropland dominates (Fig. S1). Meanwhile, the increases in cloud cover reduce available radiation, leading to weakened aerosol DRF effects over the Southeast while strengthened NPP reductions in the Southwest (Fig. S9c).

3.4 Combined effects of O_3 and aerosol

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Simultaneous inclusion of the aerosol effects on the land biosphere has negligible impacts on O_3 damage. The online O_3 inhibition, which is much stronger in magnitude than the aerosol effects, shows insignificant differences relative to the offline values (10.7% vs. 10.3%). As a result, we consider O_3 and aerosol effects to be linearly additive. In the year 2010, the combined effects of O_3 and aerosols decrease total NPP in China by 0.39 to 0.80 Pg C yr^{-1} (Table 2), equivalent to 9-16% of the pollution-free NPP and 16-32% of the total anthropogenic carbon emissions (Liu et al., 2015). A dominant fraction (86% without AIE and 77% with AIE) of the reduced carbon uptake occurs in the East, where dense air pollution is co-located with high NPP (Figs 1 and 2). Independently, O_3 reduces NPP by 0.59 Pg C yr^{-1}, with a large range from 0.43 Pg C yr^{-1} for low damaging sensitivity to 0.76 Pg C yr^{-1} for high damaging sensitivity (Table 2). The sign of the aerosol effects is uncertain. Without AIE, aerosol is predicted to increase NPP by 0.2 Pg C yr^{-1}, because of regionally confined DRF effects and enhanced soil moisture (Fig. 8). With inclusion of AIE, aerosol decreases NPP by 0.2 Pg C yr^{-1}, mainly due to reduced soil moisture (Fig. S9). The uncertainty of individual simulations, calculated from the interannual climate variability, is usually smaller than that due to O_3 damage sensitivity and AIE (Table 2).

3.5 Future projection of pollution effects

Following the CLE scenario, by the year 2030, predicted summer [O_3] increases by 7%, while AOD decreases by 5% and surface $PM_{2.5}$ concentrations decline by 10% (Fig. 9). These changes are predominantly attributed to changes in anthropogenic emissions, as natural sources show limited changes. The reduction of AOD is related to the decreased emissions of SO_2, black carbon, and organic carbon (Fig. S2). In contrast, the enhancement of [O_3] is caused by the increased emissions of NO_x accompanied with the higher air temperature in the warmer 2030 climate. The moderate decline of aerosol loading in the 2030 CLE scenario brings benefits to land ecosystems through DRF effects (Table 2) because light scattering is often saturated in the present-day conditions due to high local AOD and regional cloud cover. Benefits from the aerosol pollution reductions

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are offset by worsening O_3 vegetation damage in the CLE future world (Fig. 10b). O_3-free ($[O_3]=0$) NPP increases by 14% in 2030 due to CO_2 fertilization and global climate change. Despite $[CO_2]$ increases from 390 ppm in 2010 to 449 ppm in 2030 in the RCP8.5 scenario (van Vuuren et al., 2011), which drives additional g_s inhibition, the future O_3-induced NPP damage in 2030 degrades to 14% or 0.67 Pg C yr^{-1} (Table 2).

The MTFR scenario reflects an ambitious and optimistic future in which there is rapid global implementation of all currently available technological pollution controls. AOD decreases by 55% and $[O_3]$ decreases by 40% for this future scenario (Fig. 9). The model projects much lower damage to NPP of only 0.12 Pg C yr^{-1} (0.28 Pg C yr^{-1} with AIE), mainly from the 40% reduction in $[O_3]$ (Fig. 10c). These damages are of similar magnitude to modeling uncertainties (Table 2). The MTFR scenario offers strong recovery of the land carbon uptake in China by 2030.

4. Discussion

4.1 Comparison with previous estimates

Previous estimates of O_3 damages over the whole China region are very limited. Two important studies, Tian et al. (2011) and Ren et al. (2011), have quantified the impacts of surface O_3 on carbon assimilation in China. Both studies applied the dynamic land ecosystem model (DLEM) with O_3 damage scheme proposed by Felzer et al. (2004), except that Tian et al. (2011) focused on NEE while Ren et al. (2011) also investigated NPP. The Felzer et al. (2004) scheme calculates O_3 uptake based on stomatal conductance and the AOT40 (accumulated hourly O_3 dose over a threshold of 40 ppb). Yue and Unger (2014) estimated O_3-induced reductions in GPP over U.S. using Sitch et al. (2007) scheme and found an average value of 4-8% (low to high sensitivity), consistent with the reduction of 3-7% in Felzer et al. (2004). For this study, we estimate that present-day O_3 decreases NPP by 0.43-0.76 Pg C yr^{-1}, higher than the 0.42 Pg C yr^{-1} calculated by Ren et al. (2011). However, the percentage reduction of 10.1-17.8% in our estimate is weaker than the value of 24.7% in Ren et al. (2011). The main reason for such

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discrepancy lies in the differences in the climatological NPP. Combining all environmental drivers (e.g. [CO_2], meteorology, [O_3], and aerosols), we predict an average NPP of 3.98 ± 0.1 Pg C yr^{-1} for the year 2010 (uncertainties from AIE) with the ModelE2-YIBs model. This value is close to the average of 3.35 ± 1.25 Pg C yr^{-1} for 1981-2000 calculated based on 54 estimates from 33 studies (Shao et al., 2016). Using DLEM, Ren et al. (2011) estimated an optimal NPP of 1.67 Pg C yr^{-1} for 2000-2005 over China, which is only half of the literature-based estimate.

In the absence of any previous studies of aerosol pollution effects on land carbon uptake in China, our strategy is to compare separately the simulated aerosol climatic feedback (climate sensitivity) and simulated NPP response to climate variability (NPP sensitivity) with existing published results. ModelE2-YIBs simulates an annual reduction of 26.2 W m^{-2} in all-sky surface solar radiation over the East due to aerosols pollution (Table S6), similar to the estimate of 28 W m^{-2} by Folini and Wild (2015). In response to this radiative perturbation, aerosol pollution causes a widespread cooling of 0.3-0.9 °C in summer over the East (Fig. 7a), consistent with estimates of 0-0.9 °C by Qian et al. (2003), 0-0.7 °C by Liu et al. (2009), and average of 0.5 °C by Folini and Wild (2015). Aerosol pollution effects on regional precipitation patterns in China are not well understood due to different climate model treatments of land-atmosphere interactions and the interplay between regional and large-scale circulation. In ModelE2-YIBs, without AIE, aerosol induces a “northern drought and southern flood” pattern in agreement with Gu et al. (2006), but different to Liu et al. (2009) who predicted widespread drought instead. Including both aerosol direct and indirect effects, ModelE2-YIBs simulates an average reduction of 0.48 mm day^{-1} in summer rainfall widespread over China (Fig. S8b), similar to the magnitude of 0.4 mm day^{-1} estimated with the ECHAM5-HAM model (Folini and Wild, 2015), but higher than the 0.21 mm day^{-1} predicted by the RegCM2 model (Huang et al., 2007).

Sensitivity experiments with YIBs show that summer NPP increases following aerosol-induced changes in temperature, radiation, and precipitation (Fig. 8). The cooling-related NPP enhancement (Fig. 8b) collocates with changes in temperature (Fig. 7a), indicating that sensitivity of NPP to temperature is negative over eastern China. Such temperature sensitivity is consistent with the ensemble estimate based on 10 terrestrial models (Piao et al., 2013). For the aerosol-induced radiative perturbation, many studies have shown that moderate aerosol/cloud amount promotes plant photosynthesis through enhanced DRF, while dense aerosol/cloud decreases carbon uptake due to light extinction (Cohan et al., 2002; Gu et al., 2003; Rocha et al., 2004; Alton, 2008; Knohl and Baldocchi, 2008; Mercado et al., 2009; Jing et al., 2010; Bai et al., 2012; Cirino et al., 2014; Strada et al., 2015). Theoretically, at each specific land location on the Earth, there is an AOD threshold below which aerosol promotes local NPP. The threshold is dependent on latitude, cloud/aerosol amount, and plant types. In a related study by Yue and Unger (2016), we applied a well-validated offline radiation model to calculate these AOD thresholds over China. We conclude that present-day AOD is lower than the local thresholds in the Northeast and Southeast but exceeds the thresholds in the North China Plain, explaining why aerosol-induced diming enhances NPP in the former regions but reduces NPP in the latter (Fig. 8c). On the country level, the NPP enhancement due to aerosol DRF is 0.07 Pg C yr^{-1} in Yue and Unger (2016), very close to the 0.09 Pg C yr^{-1} estimated with ModelE2-YIBs model (Table 2).

4.2 Uncertainties

A major source of uncertainty originates from the paucity of observations. For instance, direct measurements of aerosol pollution effects on NPP are non-existent for China. The aerosol effects involve complex interactions that challenge the field-based validation of the underlying independent processes. Field experiments of O_3 vegetation damage are becoming more available, but their applications are limited by the large variations in the species-specific responses (Lombardozzi et al., 2013), as well as the discrepancies in the treatments of [O_3] enhancement (Wittig et al., 2007). Instead of equally using all individual records from multiple literatures (Lombardozzi et al., 2013), we aggregate O_3 damage for each literature based on the seasonal (or growth-season) average. In this way, the derived PFT-level GPP-[O_3] relationships are not biased towards the experiments with a large number of samplings. Such aggregation also reduces sampling noise and allows construction of the quantified GPP-[O_3] relationships used for model assessment.

We have estimated O_3 damages to NPP (instead of GPP), an optimal indicator for net carbon uptake by plants. Our calculations assume no impacts of O_3 on autotrophic respiration. Yet, limited observations have found increased plant respiration in response to O_3 injury (Felzer et al., 2007), suggesting that our calculation of O_3-induced NPP reductions might be underestimated. Current large mechanistic uncertainties in the role of anthropogenic nitrogen (N) deposition to China's land carbon uptake (Tian et al., 2011; Xiao et al., 2015) have prohibited the inclusion of dynamic carbon-nitrogen coupling in the Earth system model used here. Previous studies have suggested that inclusion of N fertilization can relieve or offset damages by O_3, especially for N-limited forests (Ollinger et al., 2002). Relative to the present day, atmospheric reactive N deposition increases by 20% in the CLE scenario but decreases by 60% in the MTFR scenario, suggesting that the stronger O_3 damage in CLE might be overestimated while the reduced damage in MTFR might be too optimistic. Furthermore, the relatively coarse resolution of the global model, usage of emission inventories, selection of aerosol parameters, and application of AIE may introduce additional biases and exacerbate the total uncertainties.

Importantly, our estimate of NPP response to aerosol effects, with or without AIE, is secondary in magnitude compared with the O_3 vegetation damage, suggesting that the net impact of current air pollution levels in China is detrimental to the land carbon uptake there. Locally, this pollution damage exerts a threat to plant health, terrestrial ecosystem services, and food production. Globally, air pollution effects may enhance planetary warming by decreasing the land removal rate of atmospheric CO_2. Our results show substantial benefits to the protection of plant health and the regional land carbon sink in China from stringent air pollution controls, especially for O_3 precursors. Our analysis highlights the complex interplay between immediate and more local pollution issues, and longer-term global warming. Future air pollution controls provide an additional co-benefit to human society: the offsetting of fossil fuel CO_2 emissions through enhanced land sequestration of atmospheric CO_2.

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Table 1. Summary of models and simulations

Model Name	Model class	Climate drivers	Number of runs	Table index [a]	Purpose
ModelE2-YIBs	Coupled climate model	Online	24	S2	Calculate ΔNPP by O_3 and aerosols at 2010 and 2030
YIBs	Vegetation model	MERRA	15	S3	Evaluate O_3 damage scheme for China PFTs
YIBs	Vegetation model	ModelE2 -YIBs	30	S4	Isolate aerosol individual climatic impacts on NPP

[a] Table index refers to the tables in supporting information.

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Table 2. Changes in NPP over China due to combined and separate effects [a] of air pollution (units: Pg C yr^{-1})

	2010	2030 CLE	2030 MTFR
O_3 (mean) [b]	**-0.59 ± 0.11 (-0.60 ± 0.13)**	**-0.67 ± 0.11 (-0.71 ± 0.16)**	**-0.29 ± 0.14 (-0.31 ± 0.10)**
Low sensitivity	-0.43 ± 0.12 (-0.40 ± 0.13)	-0.43 ± 0.14 (-0.51 ± 0.16)	-0.22 ± 0.17 (-0.15 ± 0.10)
High sensitivity	-0.76 ± 0.15 (-0.80 ± 0.16)	-0.90 ± 0.13 (-0.92 ± 0.18)	-0.36 ± 0.16 (-0.46 ± 0.12)
Aerosol (total) [c]	**0.20 ± 0.08 (-0.20 ± 0.09)**	**0.23 ± 0.14 (-0.09 ± 0.19)**	**0.16 ± 0.14 (0.04 ± 0.17)**
Temperature [d]	0.03 ± 0.04 (0.01 ± 0.04)	0.04 ± 0.02 (0.02 ± 0.05)	0.03 ± 0.04 (0.00 ± 0.04)
Radiation [d]	0.09 ± 0.04 (-0.03 ± 0.04)	0.16 ± 0.06 (-0.01 ± 0.06)	0.11 ± 0.04 (-0.03 ± 0.03)
Soil moisture [d]	0.07 ± 0.07 (-0.19 ± 0.10)	0.01 ± 0.09 (-0.09 ± 0.15)	0.03 ± 0.12 (0.00 ± 0.09)
O_3 + aerosol (net)	**-0.39 ± 0.12 (-0.80 ± 0.11)**	**-0.43 ± 0.12 (-0.80 ± 0.10)**	**-0.12 ± 0.13 (-0.28 ± 0.14)**

[a] Results shown are the averages ± one standard deviation. Simulations with both aerosol direct and indirect radiative effects (AIE) are shown in the brackets.
[b] Uncertainties for mean O_3 damages are calculated as half of differences in ΔNPP between low and high sensitivities.
[c] Combined aerosol effects are calculated with the ModelE2-YIBs climate model.
[d] Separate aerosol effects are calculated with the offline YIBs vegetation model driven with forcings from the climate model (Table S4).

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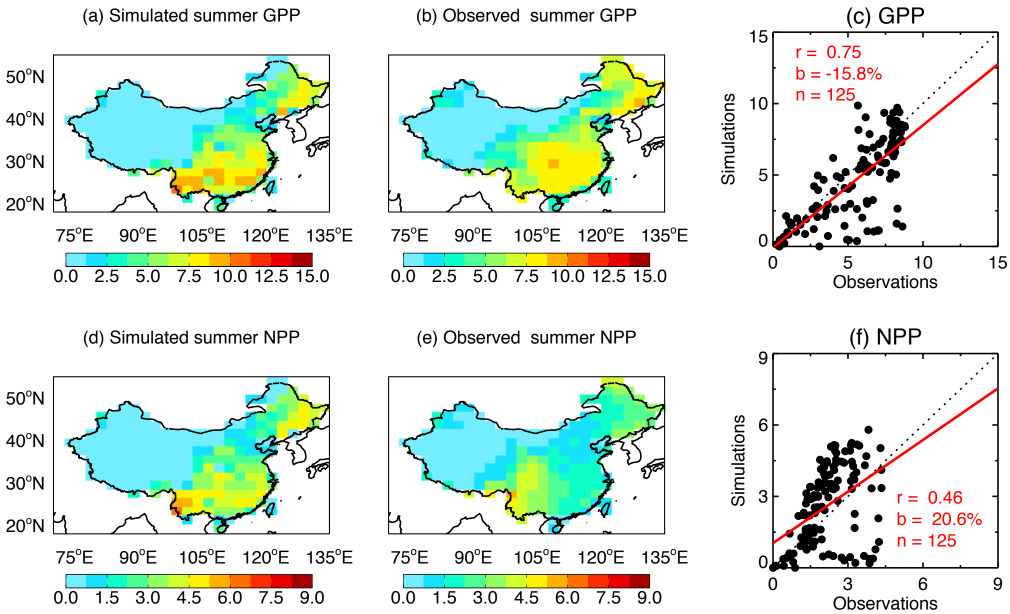

Figure 1. Evaluation of simulated summertime carbon fluxes by ModelE2-YIBs. Panels show GPP (top row) and NPP (bottom row) over China. Simulation results (a, d) are the average of G10ALLHO3 and G10ALLLO3, which are performed with the climate model ModelE2-YIBs using high and low ozone damage sensitivities (Table S2). The correlation coefficients (r), relative biases (b), and number of grid cells (n) for the comparisons are listed on the scatter plots. Units: g C m^{-2} day^{-1}.

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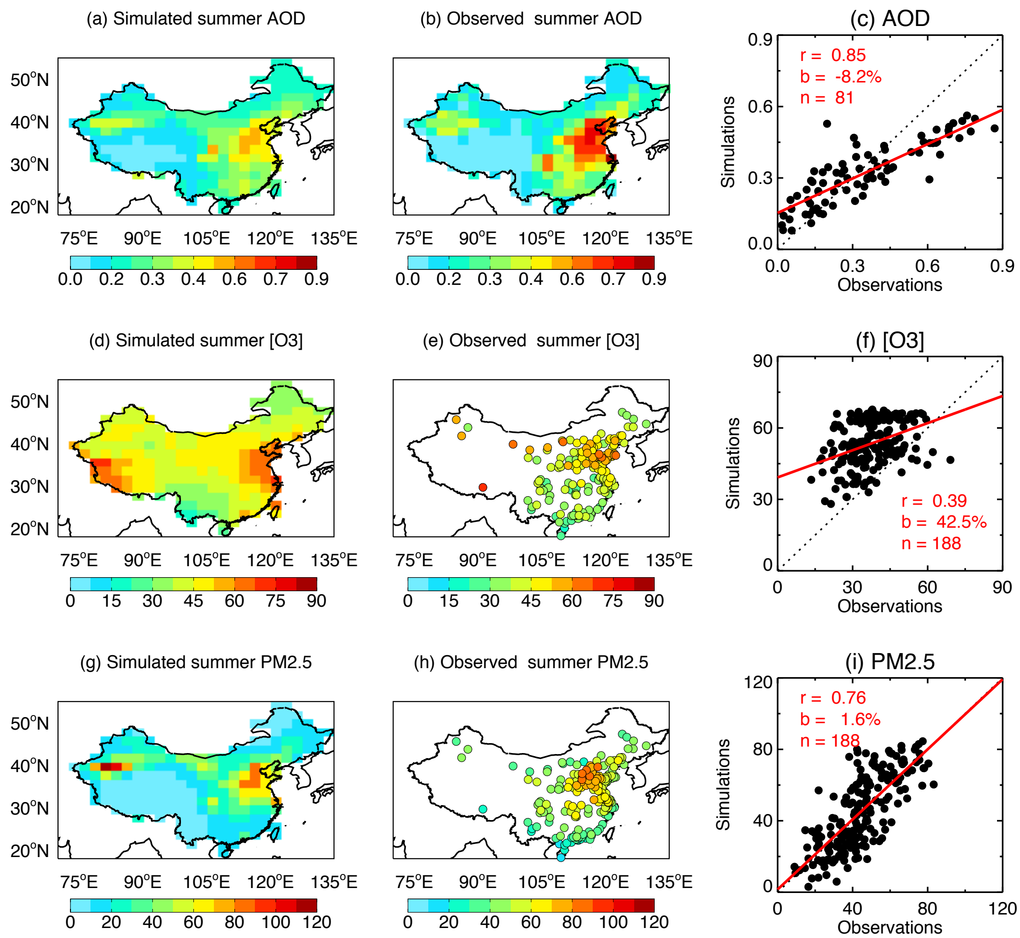

Figure 2. Evaluation of simulated summertime air pollution in China. Evaluations shown include (a) aerosol optical depth (AOD) at 550 nm, (d) [O_3] (units: ppbv), and (g) PM2.5 concentrations (units: µg m^{-3}) with observations from (b) the satellite retrieval of the MODIS, and (e) and (h) measurements from 188 ground-based sites. Simulation results are from G10ALLNO3 performed with the climate model ModelE2-YIBs (Table S2). The correlation coefficients (r), relative biases (b), and number of sites/grids (n) for the comparisons are listed on the scatter plots.

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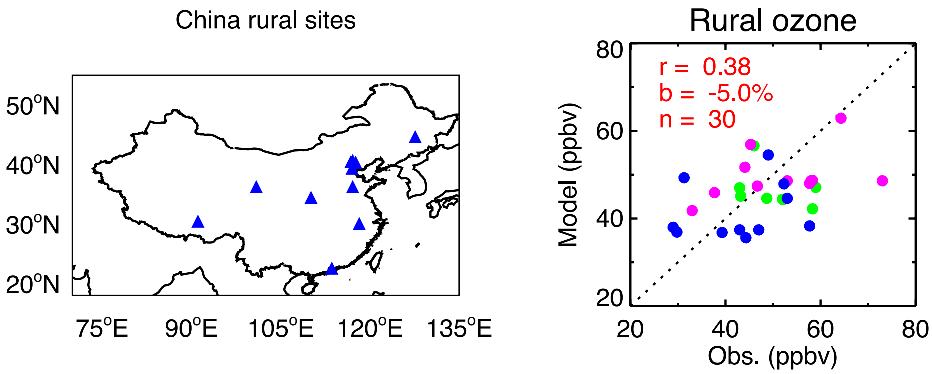

Figure 3. Evaluation of simulated [O_3] at rural sites in China. Simulation results are from G10ALLNO3 performed with the climate model NASA ModelE2-YIBs (Table S2). For the scatter plots, green, pink, and blue points represent values in spring, summer, and autumn, respectively. The data sources of all sites are listed in Table S5.

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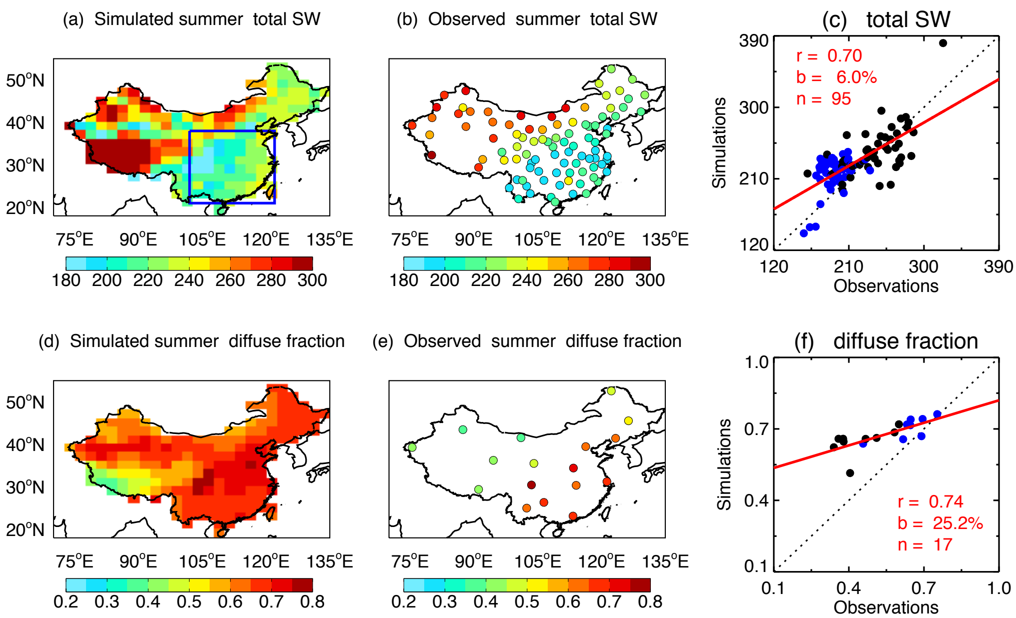

Figure 4. Evaluation of simulated radiation fluxes by ModelE2-YIBs. Panels show summertime surface (a) total shortwave radiation (units: W m^{-2}) and (d) diffuse-to-total fraction with (b, e) observations from 106 sites. Simulation results are from G10ALLNO3 performed with the climate model NASA ModelE2-YIBs (Table S2). The correlation coefficients (r), relative biases (b), and number of sites (n) for the comparisons are listed on the (c, f) scatter plots. The blue points in the scatter plots represent sites located within the box regions in southeastern China as shown in (a).

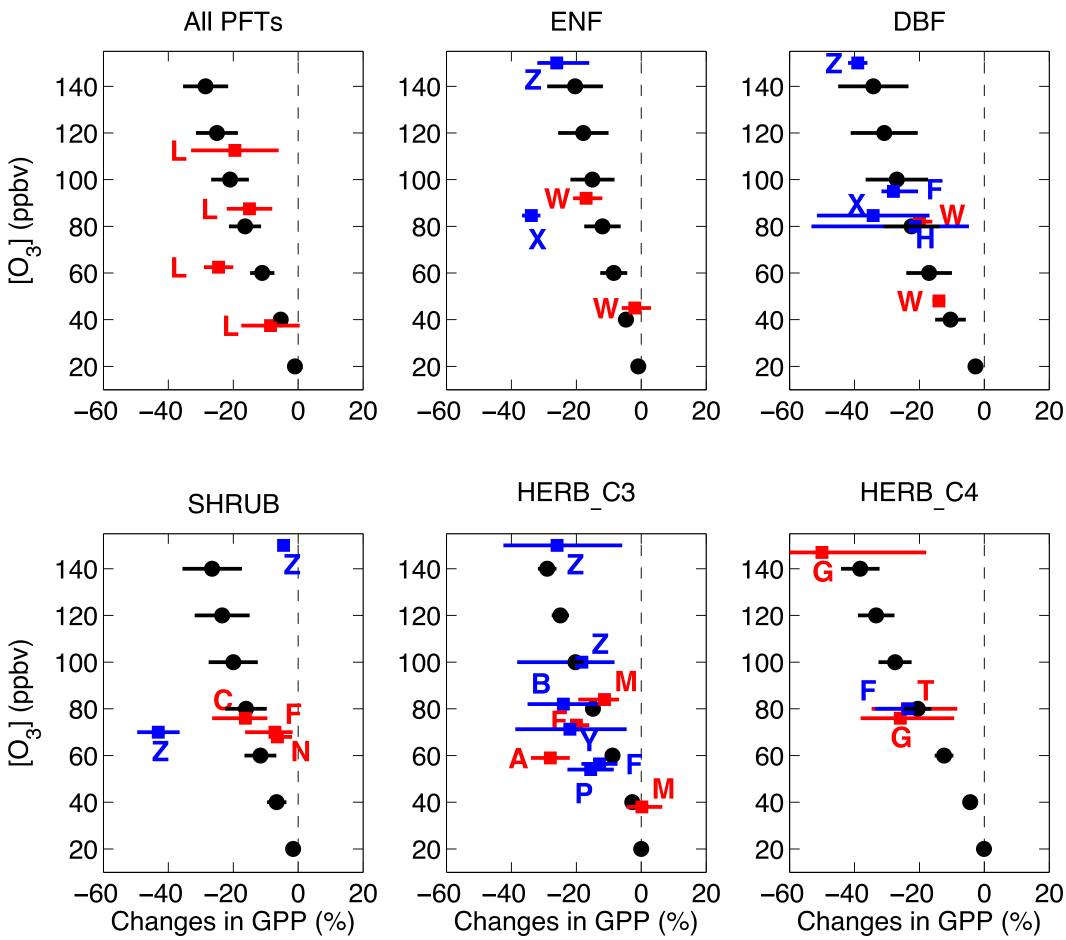

Figure 5. Comparison of predicted changes in summer GPP by O_3 with measurements. Simulations are performed using the offline YIBs vegetation model (Table S3) and averaged for all grid squares over China weighted by the area of a specific PFT. Black points show the simulated mean reductions with error bars indicating damage range from low to high O_3 sensitivity. Solid squares with error bars show the results (mean plus uncertainty) based on measurements reported in the literature (Table S1). Experiments performed for vegetation types in China are denoted with blue symbols. The author initials are indicated for the corresponding studies.

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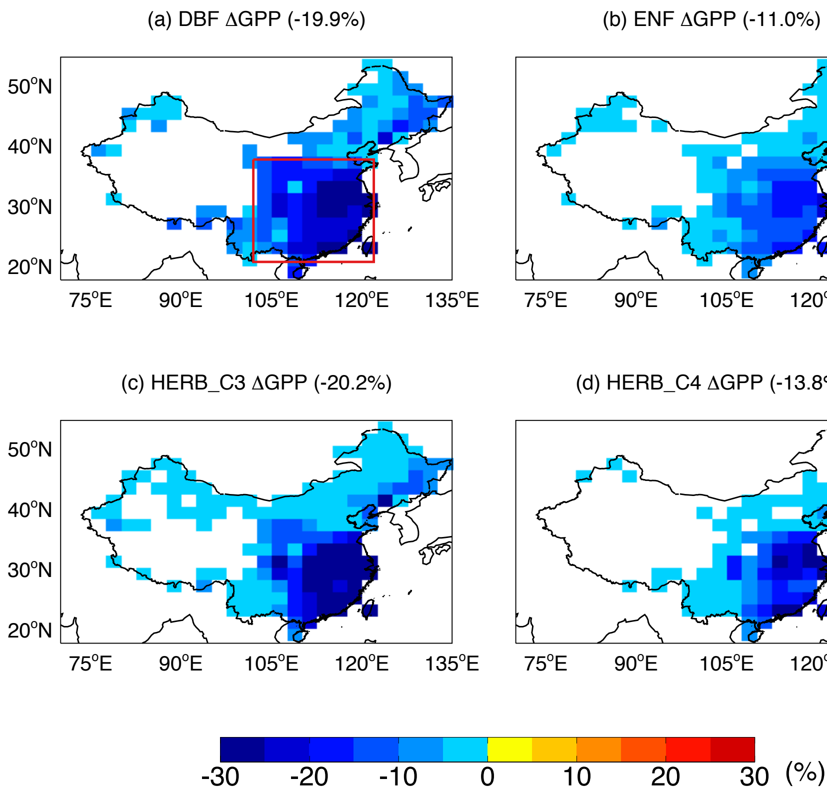

Figure 6. Predicted offline percentage damage to summer GPP caused by O_3. Panels show the damages to (a) ENF (evergreen needleleaf forest), (b) DBF (deciduous broadleaf forest), (c) C3 herbs, and (d) C4 herbs over China in the year 2010. Simulations are performed with the climate model ModelE2-YIBs, which does not feed O_3 vegetation damages back to affect biometeorology, plant growth, and ecosystem physiology. The results are averaged for the low and high damaging sensitivities:

$$(\tfrac{1}{2}(\mathrm{G10ALLHO3\_OFF}+\mathrm{G10ALLLO3\_OFF})/\mathrm{G10ALLNO3}-1)\times 100\%$$

The average value over the box domain of (a) is shown in the title bracket of each subpanel.

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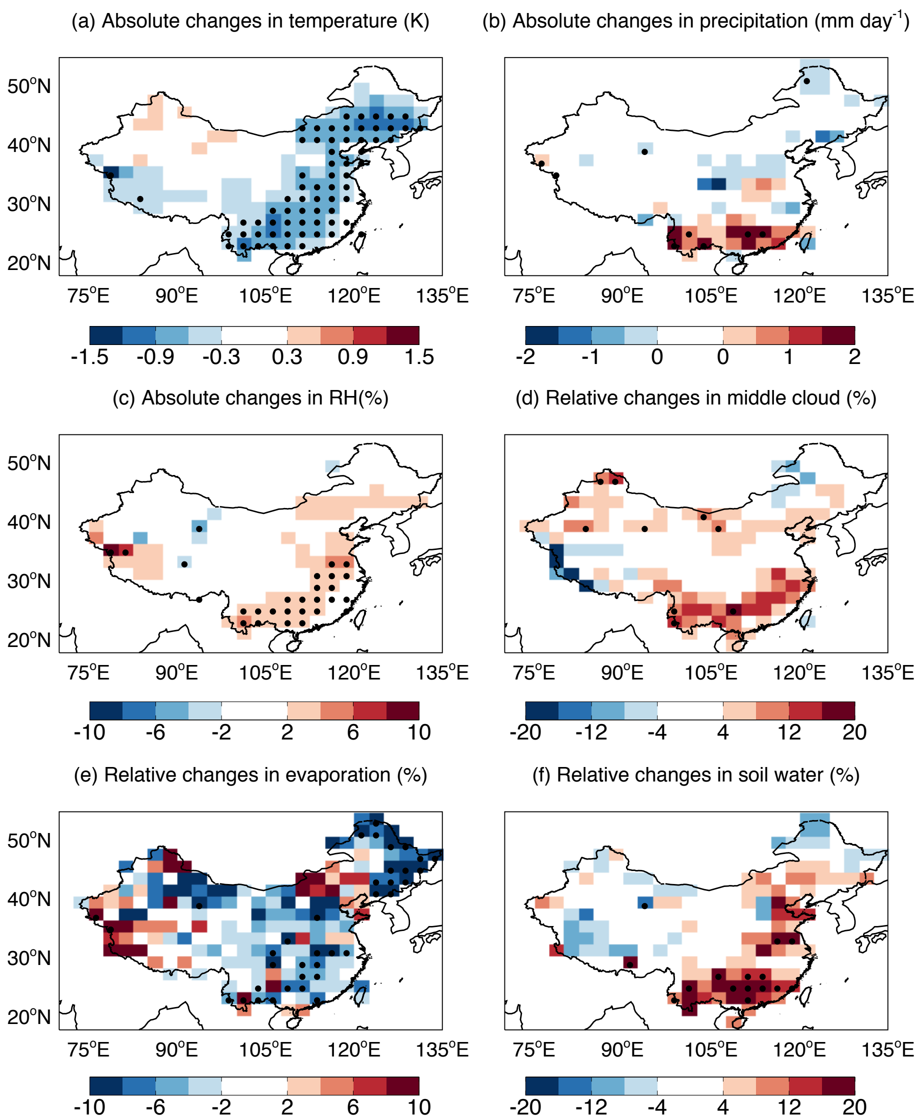

Figure 7. Changes in summer meteorology due to direct radiative effects of anthropogenic aerosols. All changes are calculated as the differences between the simulations G10ALLNO3 and G10NATNO3. For (a) temperature, (b) precipitation, and (c) relative humidity, we show the absolute changes as G10ALLNO3 – G10NATNO3. For (d) middle cloud cover, (e) evaporation, and (f) soil water content, we show the relative changes as (G10ALLNO3/G10NATNO3 – 1) × 100%. Significant changes (p<0.05) are marked with black dots.

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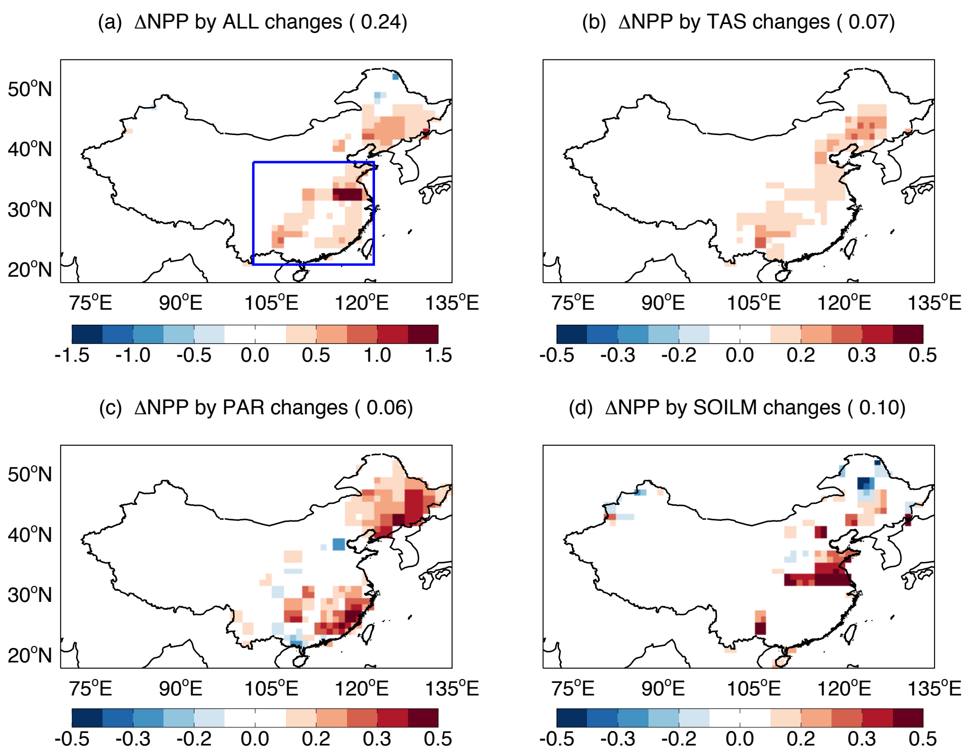

Figure 8. Decomposition of aerosol-induced changes in summer NPP. Changes in NPP are caused by aerosol-induced changes in (b) surface air temperature, (c) photosynthetically active radiation (PAR), (d) soil moisture, and (a) the combination of above three effects. Simulations are performed with the offline YIBs vegetation model driven with meteorological forcings simulated with the ModelE2-YIBs climate model (Table S4). The color scale for the first panel is different from the others. The average NPP perturbation over the box domain in a is shown in the bracket of each title. Units: g C m^{-2} day^{-1}.

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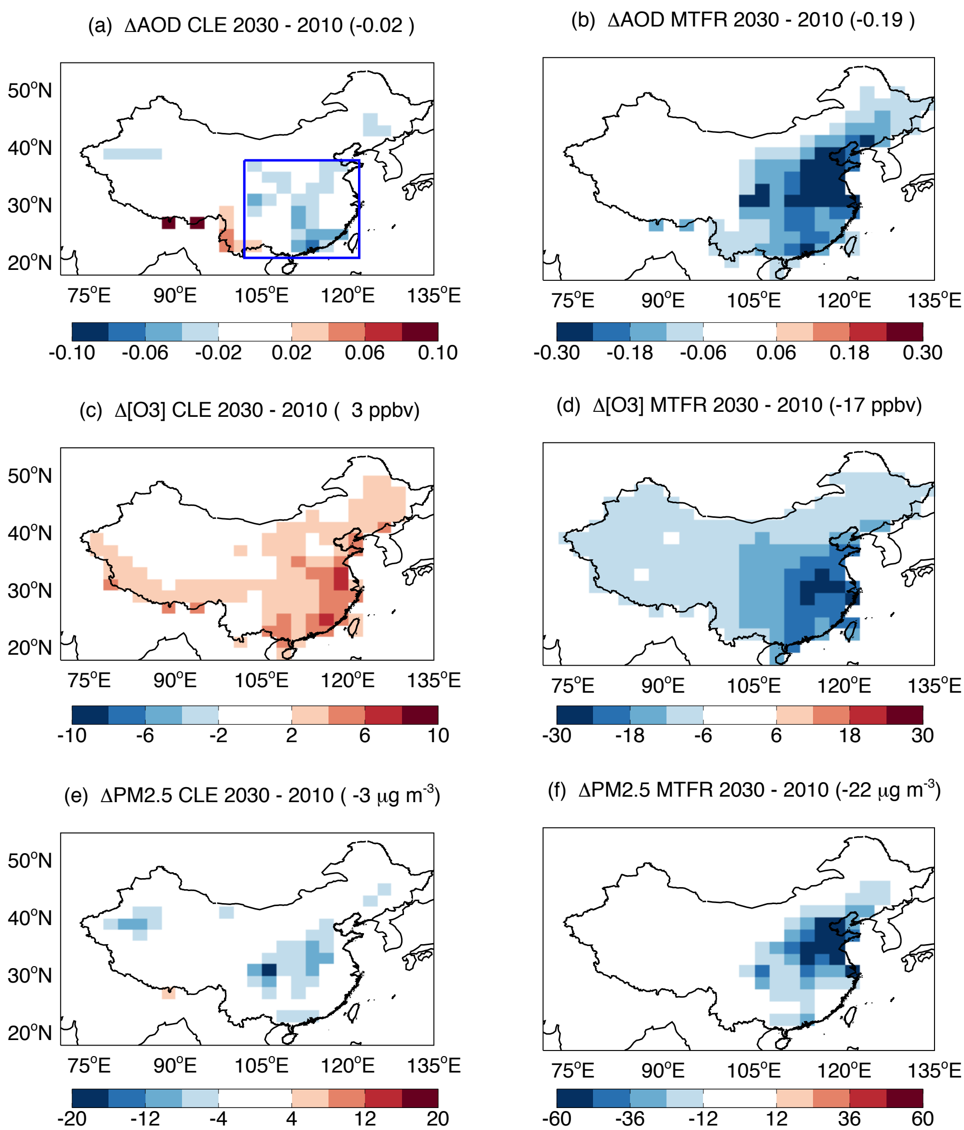

Figure 9. Predicted changes in summertime air pollution by 2030. Panels shown are for (a, b) AOD, (c, d) $[O_3]$, and (e, f) PM2.5 concentrations for the year 2030 relative to 2010 based on scenarios of (left) current legislation emissions (CLE) and (right) maximum technically feasible reduction (MTFR). Results for the left panels are calculated as (G30ALLNO3 – G10ALLNO3). Results for the right panels are calculated as (M30ALLNO3 – G10ALLNO3). The average value over the box domain of (a) is shown in the title bracket of each subpanel.

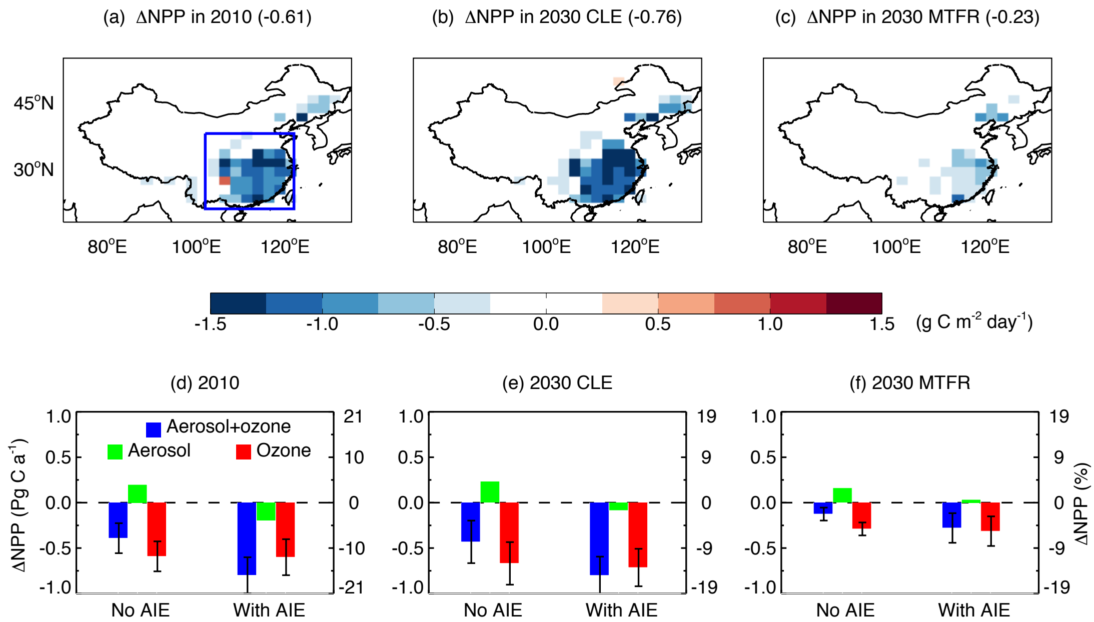

Figure 10. Impacts of air pollution on NPP in the whole of China. Results shown are combined effects of aerosol and O_3 on the summer NPP in (a) 2010, (b) 2030 with CLE scenario, and (c) 2030 with MTFR scenario. Results for the top panels do not include aerosol indirect effects (AIE) but do include the meteorological response to aerosol direct radiative effects. The average NPP perturbation over the box domain in (a) is shown in the bracket of each title. The perturbations to annual total NPP by aerosol, O_3, and their sum over the whole China are shown in (d-f) for different periods, with (right) and without (left) inclusion of AIE. Damages by O_3 are averaged for low and high sensitivities with error bars indicating ranges. The percentage changes are calculated based on NPP without AIE. Simulations are performed with the ModelE2-YIBs model. Only the significant changes ($p < 0.05$) are shown in (a-c).