**Impacts of climate and land cover changes on tropospheric ozone air quality and public health in East Asia between 1980 and 2010**

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## Abstract

Understanding how historical climate and land cover changes have affected tropospheric ozone in East Asia would help constrain the large uncertainties associated with future East Asian air quality projections. We perform a series of simulations using a global chemical transport model driven by assimilated meteorological data and a suite of land cover and land use data to examine the public health effects associated with changes in climate, land cover, land use, and anthropogenic emissions between the 5-year periods 1981-1985 and 2007-2011 in East Asia. We find that between these two periods land cover change alone could lead to a decrease in summertime surface ozone by up to 4 ppbv in East Asia and ~2000 fewer ozone-related premature deaths per year, driven mostly by enhanced dry deposition resulting from climate- and $CO_2$-induced increase in vegetation density, which more than offsets the effect of reduced isoprene emission arising from cropland expansion. Climate change alone could lead to an increase in summertime ozone by 2-10 ppbv in most regions of East Asia and ~6000 more premature deaths annually, mostly attributable to warming. The combined impacts (-2 to +12 ppbv) show that while the effect of climate change is more pronounced, land cover change could offset part of the climate effect and lead to a previously unknown public health benefit. While the changes in anthropogenic emissions remain the largest contributor to deteriorating ozone air quality in East Asia over the past 30 years, we show that climate change and land cover changes could lead to a substantial modification of ozone levels, and thus should come into consideration when formulating future air quality management strategies. We also show that the sensitivity of surface ozone to land cover change is more dependent on dry deposition than on isoprene emission in most of East Asia, leading to ozone responses that are quite distinct from that in North America, where most ozone-vegetation sensitivity studies to date have been conducted.


Keywords: climate change, land cover and land use, ozone, health impact, East Asia

## 1. Introduction

Air pollution is one of the most pressing environmental and public health concerns that we have to face today especially in rapidly developing regions such as East Asia. Several projection studies have suggested the important roles of climate and land cover changes on future air quality in addition to changing anthropogenic emissions (Fiore et al., 2012; Wu et al., 2012; Tai et al., 2013), albeit with great uncertainties not only in the projected emissions and land use, but also in the coupling between climate, atmospheric chemistry and the land cover. A better understanding of how all these factors have interacted in the past to shape air quality would be particularly useful to shed light on the likely course of atmospheric chemical evolution in the coming decades. The attribution of air quality trends and public health outcomes in East Asia, which has undergone enormous social and environmental changes over the past few decades, would also provide valuable insights for policy formulation.

One of the most important air pollutants is surface ozone ($O_3$) due to its detrimental effects on human health, and its significance in changing climate as a greenhouse gas has also been recognized (IPCC, 2013). Tropospheric ozone is produced by photochemical oxidation of precursor gases such as carbon monoxide, methane, and non-methane volatile organic compounds (VOCs) in the presence of nitrogen oxides ($NO_x \equiv NO + NO_2$). Most of these precursor gases have large anthropogenic sources, but the natural biosphere also represents significant sources depending on the region. The single most important non-methane VOC is isoprene, emitted primarily by land vegetation. Isoprene acts as a precursor for ozone in polluted, high-$NO_x$ regions, but reduces ozone by ozonolysis or by sequestering $NO_x$ as isoprene nitrate in remote, low-$NO_x$ regions. The major global sink for ozone is photolysis in the presence of water vapor, but dry deposition onto the leaf surfaces of vegetation also represents a dominant sink within the boundary layer. Surface ozone is thus dependent not only on anthropogenic emissions of precursors but also on vegetation characteristics and local chemical environments, all of which are influenced by meteorological conditions.

A strong positive correlation between temperature and ozone concentration has long been observed in many polluted regions, driven primarily by increased biogenic VOC emissions from vegetation and reduced lifetimes of peroxyacetyl nitrate (PAN) due to accelerated decomposition of PAN into $NO_x$ at higher temperatures (Jacob and Winner, 2009). This is further complicated by the covariation of temperature with frontal and cyclone passages, which represent an important ventilating mechanism for all air pollutants including ozone (Leibensperger et al., 2008; Tai et al., 2012a; 2012b). Coupled general circulation model (GCM) and chemical transport model (CTM) studies generally show that warming would lead to increased summertime surface ozone in major populated regions by 1-10 ppbv by 2050 based on IPCC future scenarios (Weaver et al., 2009; Jacob and Winner, 2009). Wang et al. (2013) reported that under the IPCC A1B scenario, climate change alone over 2000-2050 would lead to an ozone increase by up to 3.5 ppbv in eastern China but a decrease by 2 ppbv in western China, and 40% of the so-called "climate change penalty" over eastern China is attributed to enhanced biogenic VOC emissions. These studies demonstrate the potential of future climate change to at least in part offset the benefits of emission regulation in East Asia, but also highlight the large uncertainty in ozone simulations in the region arising from, e.g., different treatments of isoprene chemistry and regional chemical regimes. Few studies have examined how ozone has historically changed in East Asia, which could provide constraints for the future projections of climate change penalty on ozone air quality.

As noted above, vegetation can significantly modulate ozone air quality via biogenic VOC emissions, dry deposition, and transpiration, which controls near-surface water vapor concentration and boundary layer meteorology. Historical and future changes in the land cover and land use driven by climate change, $CO_2$ fertilization and economic activities are shown to have important ramifications for atmospheric composition. Sanderson et al. (2003) and Lathière et al. (2010) showed using a combination of vegetation models and historical land cover data that natural and anthropogenic land cover change significantly alters isoprene emission over multidecadal timescale. Wu et al. (2012) and Tai et al. (2013) predicted that changes in natural vegetation and land use over 2000-2050 could modify summertime surface ozone by up to ±5 ppbv in East Asia. Stavrakou et al. (2014) estimated that annual isoprene emission in Asia in 2005 is 21% lower if cropland expansion is accounted for in the model inputs of vegetation distribution. There are only a limited number of studies quantifying how such historical changes might have affected ozone air quality in East Asia. Fu and Liao (2014) found that seasonal mean surface ozone changes within -4 to +6 ppbv in China between the late 1980s and mid-2000s due to changes in biogenic VOC emissions driven by climate and land cover changes. This study considered only historical changes in fractional coverage of plant functional type (PFT), but not in leaf area index (LAI), dry deposition, and soil $NO_x$ emission, all of which could have large impacts on the local chemical environments and thus ozone air quality.

In this study, using historical meteorological and satellite-derived land cover data to drive a chemical transport model, we examine the individual and combined effects of changes in climate, land cover and land use on tropospheric ozone in East Asia between the 5-year periods 1981-1985 and 2007-2011, accounting for a more comprehensive set of potentially interacting mechanisms and variables including LAI (representing vegetation density), biogenic and soil $NO_x$ emissions, and dry deposition, in addition to PFT (representing vegetation distribution). We compare such effects with the contribution from anthropogenic emission changes over the same period. We further calculate the annual mortality attributable to respiratory diseases caused by ozone pollution by applying concentration-response functions from epidemiological cohort studies, as a means to explore the public health implications of historical climate change and land use trends in East Asia.

## 2. Model description and numerical experiments

### 2.1. GEOS-Chem model

We use the GEOS-Chem global 3-D chemical transport model version 9-02 (http://acmg.seas.harvard.edu/geos/), driven by assimilated meteorological data from Modern Era Retrospective-analysis for Research and Applications (MERRA) (http://gmao.gsfc.nasa.gov/merra/) with a horizontal resolution of 2.0° latitude by 2.5° longitude and reduced vertical resolution of 47 levels. MERRA, produced by the NASA Goddard Earth Observing System (GEOS), focuses on historical analysis of the hydrological cycle on a broad range of timescales and covers the modern satellite era from 1979 to present. In this work, we conduct 5-year simulations in the historical (1981-1985) and present-day (2007-2011) periods using various combinations of MERRA and land cover data. Comparisons of MERRA surface temperature (including its changes) with surface weather stations in China and NCEP/NCAR reanalysis show good agreement especially for most of the eastern half of China, reflecting a robust multidecadal trends. GEOS-Chem performs fully coupled simulations of ozone-$NO_x$-VOC-aerosol chemistry (Bey et al., 2001), and its ozone simulations over East Asia have been previously evaluated with measurements from surface sites (Wang et al., 2011; He et al., 2012) and satellites (Wang et al., 2013). These studies demonstrate the ability of GEOS-Chem to reasonably reproduce the magnitude and seasonal variation of surface ozone in the region.

Global anthropogenic emissions of CO, $NO_x$ and $SO_2$ use the EDGAR3.2-FT-2000 global inventory for 2000 (Olivier et al., 2005), and that of non-methane VOCs use the RETRO monthly global inventory for 2000 (Schultz et al., 2008). Global ammonia emissions are from the GEIA inventory (Bouwman et al., 1997). Biomass burning emissions are from the GFED-3 inventory (van der Werf et al., 2010). These global inventories are then scaled to 2005 levels. In this study, anthropogenic emissions of $SO_2$, $NO_x$, and $NH_3$ in Asia are from Streets et al. (2003; 2006), and are scaled to 2005 levels. To quantify the impact of anthropogenic emission changes, emissions for $SO_2$, $NO_x$ in Asia are then scaled to 1985 levels. The scaling factors for $SO_2$ and $NO_x$ are based on economic data and energy statistics as described by van Donkelaar et al. (2008). Emission for $NH_3$ is scaled to 1980 level by a ratio derived from historical changes between 1980 and 2003 in the Regional Emission Inventory in Asia (REAS) (Ohara et al., 2007). Methane concentrations used are fixed throughout the troposphere to annual zonal mean values in four latitudinal bands and is not determined by emission inventory.

Biogenic VOC emissions are computed by the Model of Emissions of Gases and Aerosols from Nature (MEGAN) v2.1 (Guenther et al., 2006; 2012), which is embedded in GEOS-Chem. Emissions of VOC species in each grid cell, including isoprene, monoterpenes, methyl butenol, sesquiterpenes, acetone and various alkenes, are simulated as a function of canopy-scale emission factors modulated by environmental activity factors to account for changing temperature, light, leaf age and LAI. The gridded canopy-scale emission factors are determined by the weighted average of PFT-specific emission factors and PFT fraction in each grid. We use the empirical values of PFT-specific emission factors provided by Guenther et al. (2012) (Table S1 in the Supplement). Soil $NO_x$ emission follows Yienger and Levy (1995), with updates from Hudman et al. (2012). It considers biome-specific emission factors, a continuous dependence on temperature and soil moisture, the latest gridded inventory for fertilizer and manure emissions, the timing and distribution of nitrogen fertilizer based on satellite-derived seasonality, modified length and strength of pulsed nitrogen emissions, and fertilization effect of nitrogen deposition to natural soils. Wet deposition of soluble gases and aerosols follows the scheme of Liu et al. (2001). Dry deposition follows the resistance-in-series scheme of Wesely (1989), which depends on species properties, land cover types and meteorological conditions, and uses the Olson land cover classes with 76 land types (Olson, 1992) reclassified into 11 land types. Although transpiration is a potential mechanism via which the land cover affects ozone, we do not address it in this study because water vapor concentration in GEOS-Chem is prescribed from assimilated relative humidity (i.e., not computed online from evapotranspiration).

## 2.2. Land cover and land use change

To examine the impacts of historical changes in land cover and land use (LCLU) on air quality, we derive model-specific land cover inputs for East Asia between 1980 and 2010 using the Moderate Resolution Imaging Spectroradiometer (MODIS) land cover product (MCD12Q1) with the scheme of International Geosphere-Biosphere Program (IGBP) as the baseline, which has 17 land cover types including 13 vegetation classes and 4 non-vegetated land types. To ensure the self-consistency of the PFTs across the period, we assume that the definition (vegetation composition) for each PFT remains unchanged. To obtain the land cover types used in the model, we first combine the MODIS-IGBP in year 2010 with the Koppen main climate classes following Steinkamp and Lawrence (2011). A new land cover map MODIS-IGBP-Koppen in year 2010 with 23 land cover types is developed, which is required in simulating soil $NO_x$ emission. The distribution of LCLU types in 2010 are shown in Supplement Fig. S1. The method we use to reconstruct LCLU in 1980 is similar to that of Liu and Tian (2010), and is based on the MODIS-IGBP-Koppen LCLU in year 2005 (derived similarly as with 2010) as base year and applies appropriate ratios to scale up/down the 2005 data, with the sum of fractional coverages of all land types including bareland of each grid cell always constrained to unity (see Supplement Sect. S1 for details). For biogenic VOC emissions, we merge the 23 PFTs into the 5 PFTs used by MEGAN (broadleaf trees, needleleaf trees, shrubs, crops and grasses). The details for the merging scheme are shown in Supplement Table S2. For calculating dry deposition, the model uses the Olson land map with 74 land types. Hence, we assign an Olson land type to each of the 23 land types in MODIS-IGBP-Koppen that matches the best (Supplement Table S3).

To examine the historical changes in vegetation density, an LAI dataset for East Asia for 1980-2010 are obtained from a consistent long-term global LAI product derived using a quantitative fusion of MODIS (2000-2011) and Advanced Very High Resolution Radiometer (AVHRR) (1981-2000) satellite data with a resolution of half month and 8 km (Liu et al., 2012). To represent land cover change, LAIs in year 1982 and 2010 are chosen in this study because the satellite-based LAI datasets are not available for the year 1980 and early 1981, and LAIs from these years are consistent with the average over each 5-year simulation period. Monthly mean LAIs are then averaged over the fraction of land area covered by vegetation in the model grid cell following the approach of Guenther et al. (2006) and Müller et al (2008), which are then used in the calculation of biogenic VOC emissions. The impact of interannual variations of vegetation density within the 5-year period is not explicitly included in this study, but such impact on ozone is shown to be relatively small (less than 0.5 ppbv) (Fu and Liao, 2012).

### 2.3. Numerical experiments

In this study, we conduct two sets of GEOS-Chem simulations (Table 1). For each case in the first set of simulations (Simulation I), a 5-year simulation is performed. In the control simulation [*CTRL*], present-day (2007-2011) climate, land cover types (i.e., PFT fractional coverage) and LAIs are used to drive the model. In the sensitivity simulation [*SIM_LCLU*], we use the same meteorological fields as [*CTRL*] but with the historical (1981-1985) PFTs and LAIs. In [*SIM_CLIM*], we use historical climate but present-day PFTs and LAIs. In [*SIM_COMB*], we use historical climate, PFTs and LAIs. In all the four simulations above, anthropogenic emissions of ozone precursors are set at present-day levels (2005). The [*SIM_ANTH*] simulation is the same as [*CTRL*] but with historical anthropogenic emissions of ozone precursors scaled to 1985 levels.

To determine the key factors that modulate summertime (JJA) ozone concentration, we further perform a series of sensitivity experiments for a chosen year representative of each of the present-day and historical periods (Simulation II): [*CTRL_2010*], which is simply year 2010 results from the control experiment [*CTRL*], and [*SIM_PFT*], [*SIM_LAI*], [*SIM_TMP*], and [*SIM_RH*], in which we keep every variable at the present-day level but with one of land cover types, LAIs, temperature, and relative humidity, respectively, from the historical period. Results from these sensitivity simulations enable first-order estimates of the potential relative contribution from each of the variables considered.

## 3. Changes in land cover and land use between 1980 and 2010

The vegetation changes in terms of distribution and density between 1980 and 2010 in East Asia as results of environmental and anthropogenic land use changes are shown in Fig. 1. We find that the cropland fraction in northeast and most of eastern China, Korea and various other places increases by up to 20% from 1980 to 2010, often associated with deforestation. Significant cropland-to-grassland conversion and reforestation are observed in northern China (e.g., Inner Mongolia) and many parts of southwestern and southern China, likely due to the land use polices of the Chinese government such as the “Grain for Green” project (J. Liu et al., 2010). Forested areas have generally decreased where croplands have expanded, whereas reforestation in southwestern and southern China is associated with reduced coverage of all of croplands, grasslands and shrubs (Fig. 1a).

In summer (JJA), LAI values in most of East Asia have generally increased, except in some parts of Southeast Asia (Fig. 1b). The enhanced summertime LAI is likely a result of warming and $CO_2$ fertilization, which promotes plant growth as is shown by a number of vegetation modeling studies (Gonzales et al, 2008; Kaplan et al., 2012). The pattern of satellite-derived LAI changes used in this study generally agrees with the changes derived from PFT-specific LAIs simulated by these vegetation models between 1980 and 2010. The increase in summertime LAI despite significant cropland expansion in northeastern and eastern China suggests that increased foliage density of the remaining forests may have more than offset the impact of reduced forest coverage on the grid-cell scale. On the other hand, a decline in LAI is observed in most of East Asia in spring (MAM), encompassing northeastern and southern China, Korea, and Japan (Fig. 1b). Such a decline for 1981-2006 are also reported in S. Liu et al. (2010) and Sangram (2012), possibly due to the warming-induced drought stress, reduced springtime precipitation and/or changes in agricultural practices such as the earlier end of spring harvest season in semiarid drylands of India (Sangram, 2012), the clearance of forests and brushes before crop and timber production through fire burning in Southeast Asia (S. Liu et al., 2010), and structural adjustments of agriculture in eastern China (Hou et al., 2015).

## 4. Impacts of land cover and land use change alone on ozone air quality

Figures 2a and 3a show the changes in surface ozone concentration arising from 1980-2010 LCLU change alone in summer and spring, respectively, expressed as seasonal mean of maximum daily 8-hour average ozone concentration (MDA8 $O_3$). Summertime surface ozone changes by ±2 ppbv in many regions of East Asia, particularly in most of China (except in Tibet and southern China), Korea and Japan where ozone decreases locally by up to 4 ppbv (Fig. 2a). We find that such LCLU-driven decreases in ozone are primarily driven by increased summertime LAI that leads to enhanced ozone dry deposition (Fig. 2c). Much of China east of ~100 °E is in a high-

$NO_x$, VOC-limited regime. For example, in much of central China and Japan, enhanced isoprene emission should increase ozone production, but the decreases in ozone in those regions indicate that enhanced isoprene emission might play a smaller role in affecting ozone than enhanced dry deposition, which decreases ozone. Exceptions include northeastern China, where reduced isoprene emission following cropland expansion (despite increased LAI) contributes in part to the lower ozone; and southern China, where higher isoprene emission following increased forest coverage and LAI contributes significantly to the increased ozone there. In spring, an ozone increase in the range of 0.5-2 ppbv is found in most of China (except part of eastern China) and part of Southeast Asia (Fig. 3a). In much of China, Korea and Japan, the changes in springtime ozone are largely driven by dry deposition changes. Places where isoprene emission changes may be important include southwestern China and some parts of Southeast Asia, where a $NO_x$-limited regime prevails and the strong reduction in isoprene emission, together with increased soil $NO_x$ and reduced dry deposition, leads to worse ozone air quality. See Supplement Sect. S2 for details of isoprene emission changes. Our results indicate that the land use change such as cropland expansion in some regions could be beneficial for ozone air quality through reducing biogenic emissions, since crops are generally low-emitting species. However, such effects may be complicated by that some economic biofuel crops such as oil palms are high isoprene emitters, and large-scale replacement of nature vegetation with these crops is expected to increase biogenic emissions (Kesselmeier et al., 1999; Guenther et al., 2006; Wiedinmyer et al., 2006), and thereby enhancing ozone depending on the region. Although such replacement is not characteristic of the history and the regions focused in this study, future work concerning ozone-crop interactions should definitely consider the effects of different crop types.

Compared with the results of Fu and Liao (2014), our simulated impacts of LCLU on surface ozone are generally larger over China. Fu and Liao (2014) primarily considered the roles of vegetation distribution in affecting biogenic VOC emissions only. Our results demonstrate that dry deposition and vegetation density are equally, and potentially more, important in shaping ozone air quality in East Asia depending on the region. We investigate this further by considering the two important factors via which the land cover could influence ozone in the model – PFT fractional coverage (representing vegetation distribution) and LAI (representing vegetation density) – and comparing the results from [*CTRL_2010*] with those from [*SIM_PFT*] and [*SIM_LAI*] to better understand the relative importance of these two vegetation parameters. Without LAI changes, changes in PFT distribution alone reduce JJA surface ozone by up to 4 ppbv in Japan, Korea, northeastern, eastern and southwestern China, and parts of Southeast Asia, whereas in southern and western China it increases by 0.5-2 ppbv (Supplement Fig. S3a). This indicates that cropland expansion might benefit public health in VOC-limited regions due to reduced isoprene emission, but might worsen ozone air quality in low-$NO_x$ regions due to reduced isoprene and increased soil $NO_x$ emissions (Supplement Fig. S3a). Afforestation would have the opposite effects. On the other hand, as a result of LAI changes alone, JJA ozone exhibits reduction by as much as 2 ppbv in most of China (except in southern China), primarily driven by increased dry deposition following increased JJA LAI, though in part offset by increased isoprene emission in VOC-limited regions (Supplement Fig. S3b). Enhanced LAI leads to higher ozone in southern China, which is the most isoprene-abundant (but still high-$NO_x$) region of China. The LAI (density) effect generally dominates over the PFT (distribution) effect in East Asia.

## 5. Impacts of climate change alone on ozone air quality

Figure 4 shows the effects of climate change alone between 1980 and 2010 on surface ozone ([*CTRL*]-[*SIM_CLIM*]). Simulated surface ozone changes in summer are within the range of -2 to +12 ppbv over East Asia due to climate change alone, with the largest over Mongolia, eastern and northeastern China, representing significant “climate penalty” on ozone regulatory effort during the study periods (Fig. 4a). In contrast, surface ozone decreases in some parts of western China by up to 2 ppbv. Surface ozone increases in spring by as much as 8 ppbv in southern and eastern China, but decreases by up to 4 ppbv over mid-latitude regions of East Asia (~30-40 °N) and in Myanmar (Fig. 4b).

We further investigate the impact of individual meteorological variable on surface ozone by comparing the results from [CTRL_2010] with the sensitivity simulations [SIM_TMP] and [SIM_RH] (Supplement Sect. S4). Both the temperature-driven or relative humidity-driven ozone changes are consistent with the large temperature and humidity changes identified, indicating their significant roles in ozone formation and destruction. From the sensitivity simulations we find that the 1980-2010 ozone changes in most of East Asia are primarily driven by changes in temperature in both summer and spring (Fig. 4c and 4d), reflecting enhanced isoprene emission and PAN decomposition at higher temperatures in these mostly high-$NO_x$ regions (Wang et al., 2013). The widespread summertime ozone increase east of ~110°E (Fig. 4a) are also driven in lesser part by reduced relatively humidity (Fig. 4e), which inhibits ozone destruction in the presence of water vapor. The summertime ozone increase in the Shandong province of China despite a small drop in temperature and rise in relative humidity may reflect influence of westerly transport. The ozone decrease in west-central China in both seasons is also consistent with enhanced relative humidity (Fig. 4e and 4f) and reduced mixing height (Fig. 4g and 4h). This agrees with Dawson et al. (2007) who found that in the eastern US a shallower mixing height reduces ozone in polluted areas, because it increases $NO_x$ concentration in the mixed layer and thus inhibits ozone production in regions with an overabundance of $NO_x$ (Kleeman, 2008; Jacob and Winner, 2009).

## 6. Comparison between the impacts of changes in climate, land cover and anthropogenic emissions

Under the combined effects of climate and LCLU changes ([*CTRL*]-[*SIM_COMB*]) between 1980 and 2010, changes in summertime surface ozone range between -2 and +12 ppbv in most of East Asia, with an ozone enhancement of 2-8 ppbv in the most densely populated coastal regions (Fig. 5a). We also examine the interannual variations of surface ozone concentration within each 5-year period based on the simulations CTRL and COMB, which are quantified using the mean absolute deviation (MAD) (Supplement Fig. S5). We find that the interannual variations vary within the range of 0.2-3.0 ppbv across East Asia. Therefore, in comparison with such variations, the changes in surface ozone induced by climate and LCLU changes in this study are shown to be significant. The spatial pattern of ozone changes under the combined effects is similar to that from climate change alone (Fig. 4a), reflecting the more dominant role of climatic factors in affecting ozone, although the LCLU effects often offset (and at some locations enhance) a sizable portion of the climate effects. In several places (e.g., Japan and Korea), the sign of change from the combined effects even becomes opposite to that from climate change alone, indicating the importance of LCLU change in offsetting the climate-driven ozone increases in some East Asian regions.

Figure 5b shows that the changes in anthropogenic emissions alone (with fixed, present-day climate and LCLU) between 1985 and 2005 enhance summertime ozone by 2-25 ppbv in East Asia ([*CTRL*]-[*SIM_ANTH*]), reflecting the unsurprisingly dominant role of anthropogenic emissions in controlling East Asian air quality over the past few decades. The largest ozone increase occurs in southern, eastern and central China. Against the backdrop of changing emissions, Fig. 5a demonstrates that in most of East Asia, emission-driven ozone increases could be substantially enhanced by multidecadal changes in climate but then partially offset by climate- and $CO_2$-driven changes in vegetation density.

## 7. Impacts on human health

Previous epidemiological studies have shown that ozone has a detrimental effect on human health, and the exposure to ozone could lead to premature respiratory mortality (e.g., Jerrett et al., 2009). We thus further assess the possible public health implications of historical ozone changes in East Asia as a result of changes in climate, land cover, and anthropogenic emissions between the periods 1981-1985 and 2007-2011. Because there are very limited studies reporting long-term ozone-related mortality in East Asia, we apply epidemiological concentration-response functions (CRFs) from American Cancer Society (ACS) in this study following the methods of Anenberg et al. (2010) and Silva et al. (2013). The estimates of excess ozone-related respiratory mortality ($\Delta M$, in 1000 deaths per year per squared km) for all adults aged 30 and above are calculated by

$$\Delta M = y_0\left(1 - e^{-\beta \Delta X}\right)P$$

where $y_0$ represents the baseline mortality rate (deaths per thousand people per year), $\beta$ is a concentration-response factor, $\Delta X$ represents the differences in ozone concentration in terms of April-September 6-month averaged of 1-h daily maximum ozone concentration (Jerrett et al., 2009), and $P$ is the exposed population (people per squared km). Please see Supplement Sect. S6 for details.

Figure 6 shows the estimates with uncertainties of the ozone-related respiratory mortality (concerning adults aged 30 and above) attributed to historical climate change ([*CTRL*]-[*SIM_CLIM*]), land cover and land use change ([*CTRL*]-[*SIM_LCLU*]), and anthropogenic emissions ([*CTRL*]-[*SIM_ANTH*]). The mortality attributed to past changes in anthropogenic emissions is the largest - about 61 600 more deaths in East Asia and 28 370 more deaths in China annually. The effect of past climate change on mortality is 5600 more deaths and 4409 more deaths annually in all of East Asia and China, respectively. Historical LCLU change (mainly via climate- and $CO_2$-driven increase in vegetation density) causes ozone-related respiratory mortality to decrease by 2200 deaths $yr^{-1}$ in East Asia and by 243 deaths $yr^{-1}$ in China, reflecting the relatively small but not insignificant public health benefit of multidecadal LCLU change over the past 30 years due to the alleviation of ozone pollution that in part offsets the health damage of warming and increasing emissions.

## 8. Conclusions and discussion

In this study, we investigate the effects of changes in climate, land cover and land use between the periods 1981-1985 and 2007-2011 on surface ozone concentration in East Asia using the GEOS-Chem chemical transport model driven by assimilated meteorological fields, land use data from historical RCP harmonization, and several satellite-derived land cover products. We characterize the possible changes in vegetation distribution and density, as well as various climate variables, in East Asia in study periods, and examine their influences on ozone air quality and public health along the backdrop of changing anthropogenic emissions, focusing on spring and summer when ozone pollution is usually the most serious. East Asian land cover change is generally characterized by a reduction in forest coverage (mostly due to cropland expansion) in most of southern, eastern and northeastern China and adjacent regions, but an increase in forest coverage in parts of northern, southwestern and western China. LAI has generally increased in summer likely due to warming and $CO_2$ fertilization, but decreased in spring.

From the simulations using different combinations of present-day (2007-2011) vs. historical (1981-1985) meteorological fields, land cover data and anthropogenic emissions of ozone precursors, we estimate that historical land cover and land use change alone between 1980 and 2010 could have led to reduced summertime surface ozone by up to 4 ppbv in most of East Asia, driven mainly by warming- and $CO_2$-induced enhancement in summertime LAI, but enhanced springtime ozone by 0.5-2 ppbv in most of East Asia. Historical climate change alone has increased summertime surface ozone by 2-10 ppbv in most places of East Asia except in some parts of western China. In spring, climate change alone has increased surface ozone by up to 8 ppbv in southern and eastern China, but decreased ozone by as much as 4 ppbv over much of the midlatitude regions of East Asia. Such climate effects are driven mainly by changes in seasonal mean temperature. Changes in anthropogenic emissions of ozone precursors mostly from industrial sources remain the largest contributor to worse ozone air quality (by as much as 25 ppbv) in most of East Asia, but climate change could substantially further enhance ozone, while land cover and land use change could partially offset the rising ozone levels in various regions over the past 30 years. We further examine the public health implications of these results by estimating the possible changes in annual mortality attributable to ozone-related respiratory diseases between 1980 and 2010. Rising anthropogenic emissions have increased respiratory mortality by tens of thousands more deaths per year in East Asia over the past three decades. The multidecadal land cover change (mostly via enhanced vegetation density), however, might have alleviated the emission-driven health impacts, while climate change (mostly warming) might have aggravated those impacts. Such results highlight the importance of considering the effects of future climate and land cover changes in formulating adequate emission control strategies to tackle public health issues related to air pollution.

We also find that, at least in ways represented in our model, the effects of land cover change on ozone air quality in East Asia differ substantially from those elsewhere due to different background chemical environments. Changes in vegetation density and dry deposition appear to be more important factors in East Asia than changes in plant type distribution and isoprene emission, whereas the opposite is true in most of the US, which contains one of the largest isoprene hotspots in the world (Millet et al., 2008; Tai et al., 2013), even though both regions are mostly high-$NO_x$. Future work should thus focus on a more systematic analysis on the global spatial variability of ozone sensitivity to vegetation changes (e.g., driven by climate and land use changes), which may yield opposite responses depending on the region. Likewise, cropland expansion is shown to affect ozone but the sign of effect also depends on the relative importance of dry deposition vs. biogenic emissions. In addition, the replacement of natural vegetation with high isoprene-emitting species such as some biofuel crops may further complicate the effects, and the implications for air quality need to be considered in future studies especially for tropical East and Southeast Asia. Our study also does not account for the changes in manure and chemical fertilizer associated with changes in LCLU and agriculture practices (Potter et al., 2010), which could affect soil $NO_x$ emission and ozone concentration, though such effects are expected to be relatively minor given the VOC-limited regions prevalent in most of China.

Previous studies have indicated that ambient $CO_2$ level could affect isoprene emission and thus the air quality (Possell et al., 2005, 2011; Wilkinson et al., 2009), but this effect is not considered here. Tai et al. (2013) suggested that the inclusion of $CO_2$ inhibition would generally reduce the sensitivity of surface ozone to climate and natural vegetation where isoprene emission is important. However, experimental data for $CO_2$-

isoprene relationship at sub-ambient $CO_2$ levels characteristic of the past are generally scarce and not consistent enough to buttress inclusion for our model period.

Another source of uncertainty is related to the use of several independent land cover datasets, which all contain various degrees of errors and may not be consistent with one another. Though in part cross-validated with potential vegetation from dynamic vegetation models, the limited spatial and temporal information provided by existing datasets still poses a challenge for a complete characterization and physical interpretation of land cover change in many of the regions concerned. In this study, we assume the vegetation composition for each vegetation type and the resistance values for each dry deposition land type remain unchanged between 1980 and 2010. How compositional changes in each PFT in response to future environmental changes will affect air quality definitely warrants further investigation. Finally, there have been relatively few related long-term studies concerning air quality and health in East Asia, thus the health impact functions and parameters used in this study are only derived from a limited number of epidemiological cohort studies mainly in North America. More regionally specific information for the relationships between human health and long-term ozone exposure is required to constrain the estimates of the public health impacts of climate and land cover changes in future studies.

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Table 1. Summary of the simulations in this study

| Simulations | MERRA meteorology | Vegetation parameters | | Anthropogenic emissions |
|---|---|---|---|---|
| | | PFT distribution * | LAI * | |
| **Simulation I** | | | | |
| CTRL | 2007-2011 | 2010 | 2010 | 2005 |
| SIM_LCLU | 2007-2011 | 1980 | 1982 | 2005 |
| SIM_CLIM | 1981-1985 | 2010 | 2010 | 2005 |
| SIM_COMB | 1981-1985 | 1980 | 1982 | 2005 |
| SIM_ANTH | 2007-2011 | 2010 | 2010 | 1985 |
| **Simulation II** | | Only for summer (JJA) | | |
| CTRL_2010 | 2010 (*T*, RH. etc.) * | 2010 | 2010 | 2005 |
| SIM_PFT | 2010 (*T*, RH. etc.) | 1980 | 2010 | 2005 |
| SIM_LAI | 2010 (*T*, RH. etc.) | 2010 | 1982 | 2005 |
| SIM_TMP | 1982 *T*; 2010 (RH. etc.) | 2010 | 2010 | 2005 |
| SIM_RH | 1982 RH; 2010 (*T*. etc.) | 2010 | 2010 | 2005 |

*. *T*: temperature; RH: relative humidity; PFT: plant function type; LAI: leaf area index.

## Figure Captions

*Figure 1.* (a) 1980-2010 changes in fractional coverage of croplands, forests (needleleaf + broadleaf + mixed), grasslands and shrubs; (b) changes in summertime (JJA) and springtime (MAM) LAI between 1980 and 2010.

*Figure 2.* Changes in summertime (JJA) (a) surface maximum daily 8-hour average ozone concentration (MDA8 $O_3$); (b) isoprene emission; (c) ozone dry deposition velocity; and (d) soil $NO_x$ emission, driven by 1980-2010 changes in land cover and land use alone ([*CTRL*] – [*SIM_LCLU*]). Values are differences between the five-year averages over the present-day and historical periods.

*Figure 3.* Changes in springtime (MAM) (a) surface maximum daily 8-hour average ozone concentration (MDA8 $O_3$); (b) isoprene emission; (c) ozone dry deposition velocity; and (d) soil $NO_x$ emission driven by 1980-2010 changes in land cover and land use alone ([*CTRL*] – [*SIM_LCLU*]). Values are differences between the five-year averages over the present-day and historical periods.

*Figure 4.* Changes in (a) surface maximum daily 8-hour average ozone concentration (MDA8 $O_3$) in summer (JJA); (b) surface MDA8 $O_3$ in spring (MAM); (c) mean JJA temperature; (d) mean MAM temperature; (e) mean JJA relative humidity; (f) mean MAM relative humidity; (g) mean JJA planetary boundary layer (PBL); and (h) mean MAM PBL driven by 1980-2010 changes in climate alone ([*CTRL*] – [*SIM_CLIM*]). Values are differences between the five-year averages over the present-day and historical periods.

*Figure 5.* Changes in summertime (JJA) surface maximum daily 8-hour average ozone concentration (MDA8 $O_3$) driven by changes in (a) climate, land cover and land use combined ([*CTRL*] – [*SIM_COMB*]); and (b) anthropogenic emissions alone ([*CTRL*] – [*SIM_ANTH*]).

*Figure* 6. Estimates of ozone-related respiratory mortality (in 1000 deaths $yr^{-1}$) attributable to historical (1980-2010) changes in land cover and land use (LCLU), climate (CLIM), climate and LCLU combined (COMB), and anthropogenic emissions (ANTH) in all of East Asia and China. Uncertainty for each case represents the 95% confidence interval of the concentration-response function.

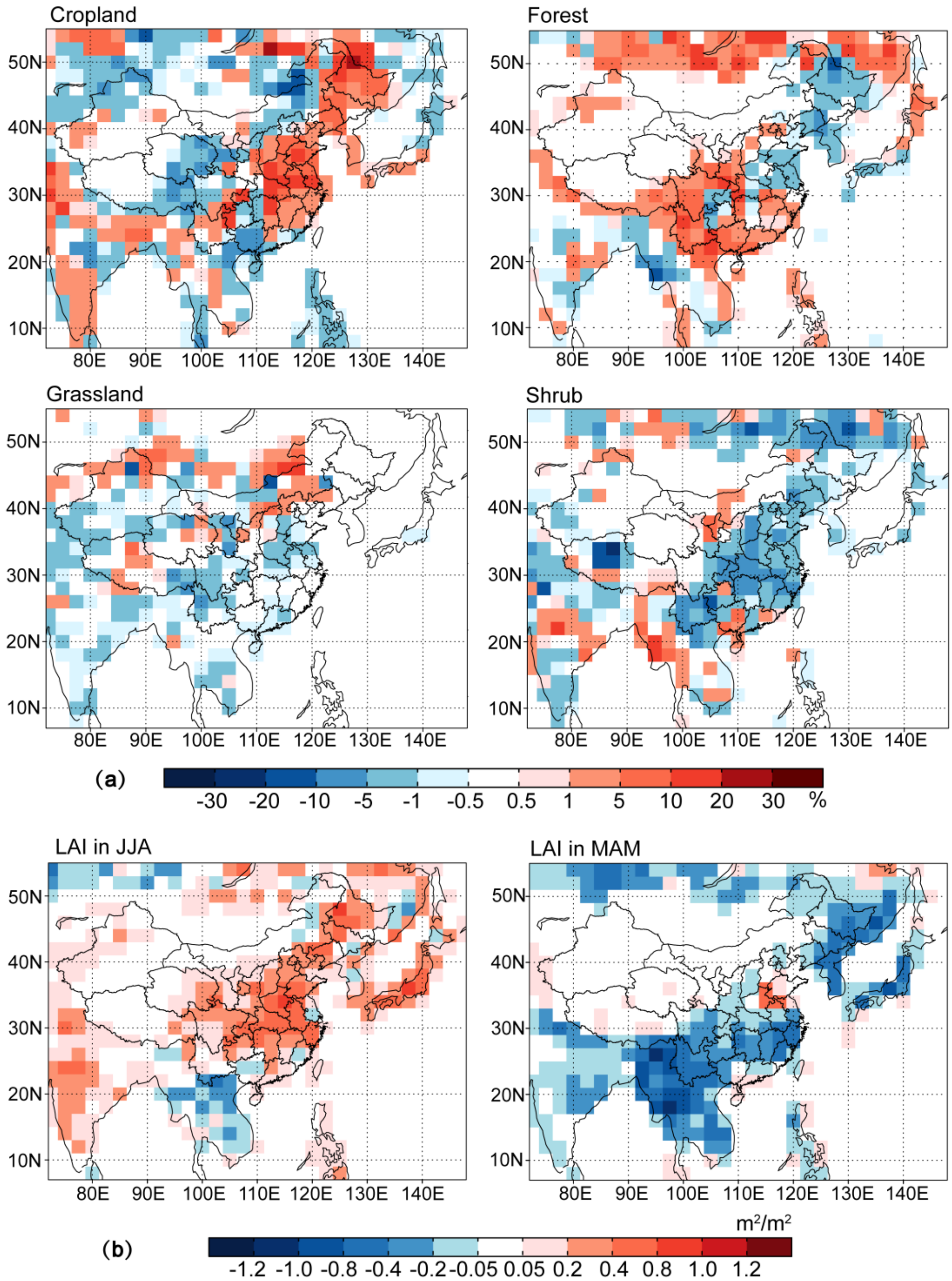


Figure 1. (a) 1980–2010 changes in fractional coverage of croplands, forests (needleleaf+broadleaf+mixed), grasslands and shrubs; (b) changes in summertime (JJA) and springtime (MAM) LAI between 1980 and 2010.

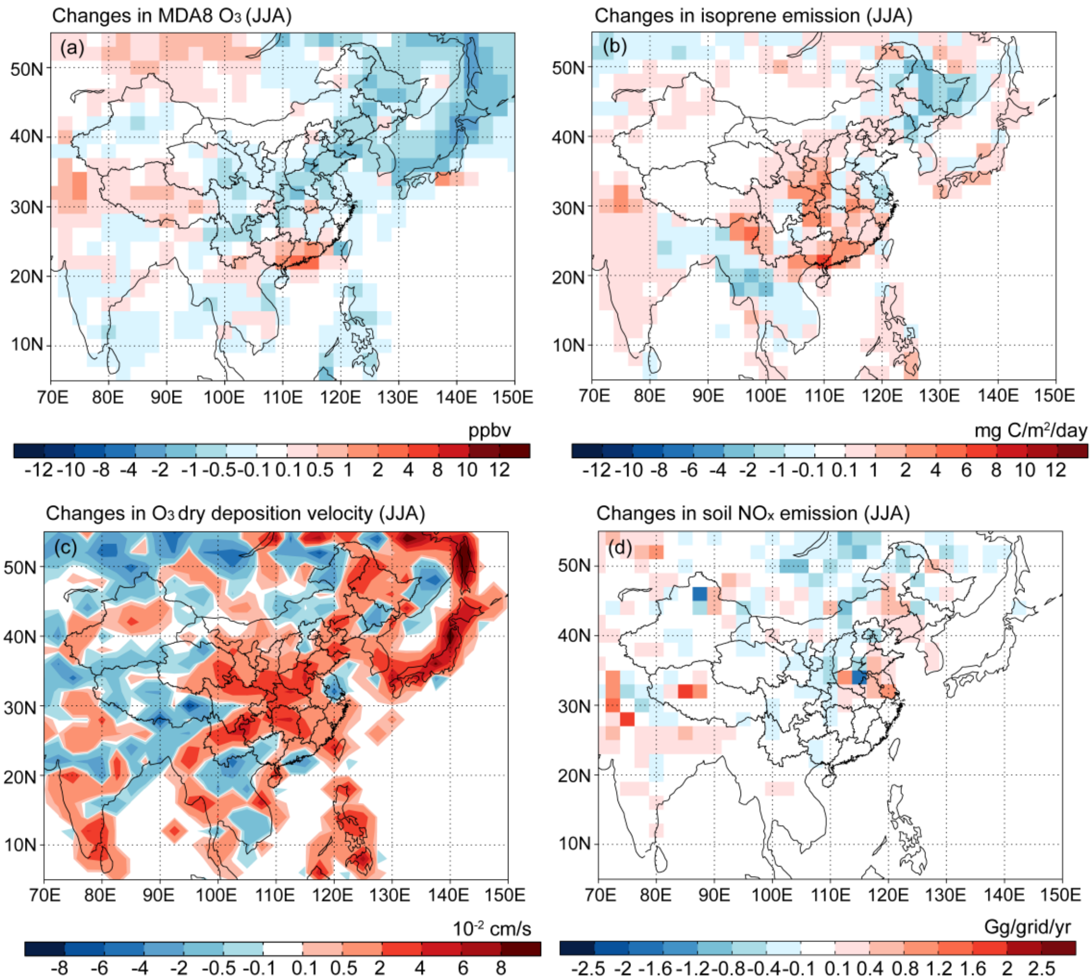


Figure 2. Changes in summertime (JJA) (a) surface maximum daily 8 h average ozone concentration(MDA8 $O_3$); (b) isoprene emission; (c) ozone dry deposition velocity; and (d) soil $NO_x$ emission, driven by 1980–2010 changes in land cover and land use alone ([CTRL]–[SIM_LCLU]). Values are differences between the five-year averages over the present-day and historical periods.

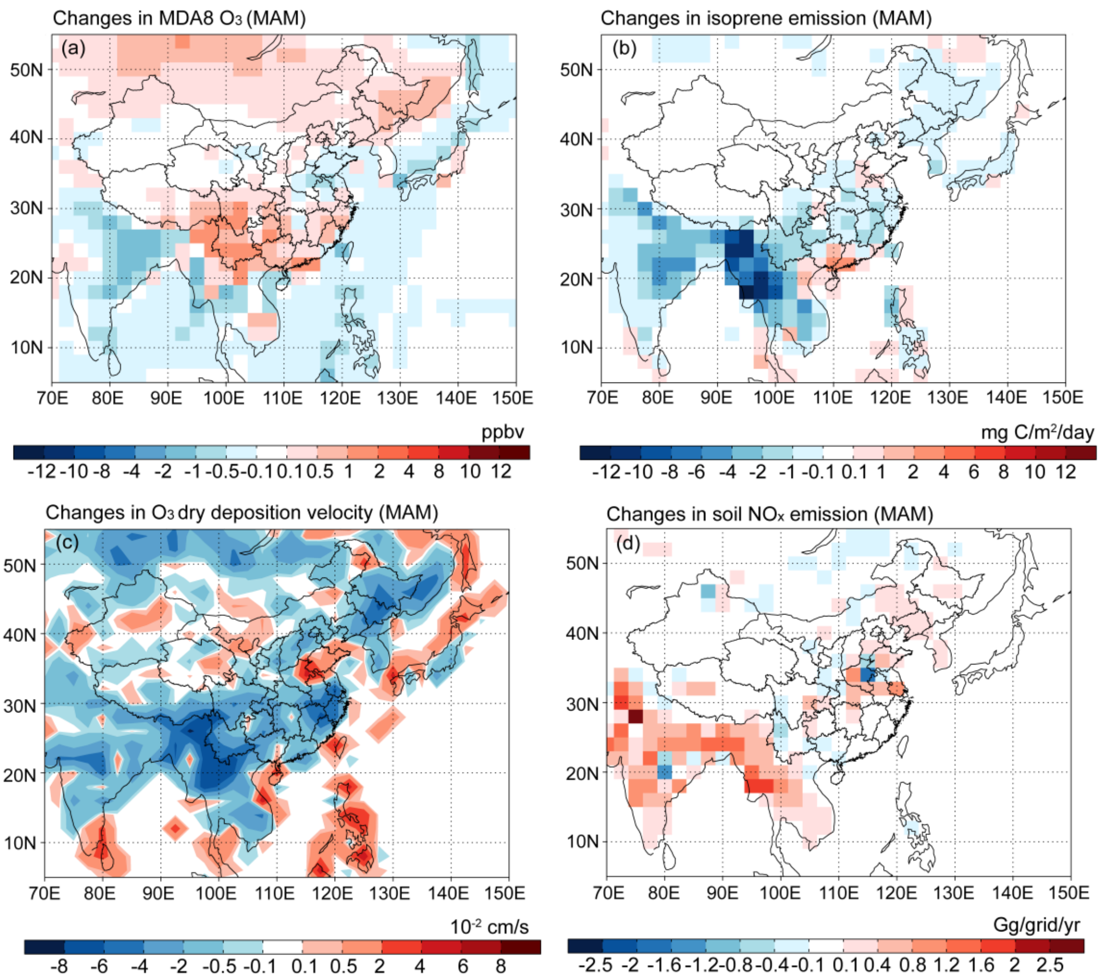


Figure 3. Changes in springtime (MAM) (a) surface maximum daily 8 h average ozone concentration (MDA8 $O_3$); (b) isoprene emission; (c) ozone dry deposition velocity; and (d) soil $NO_x$ emission driven by 1980–2010 changes in land cover and land use alone ([CTRL]–[SIM_LCLU]). Values are differences between the five-year averages over the present-day and historical periods.

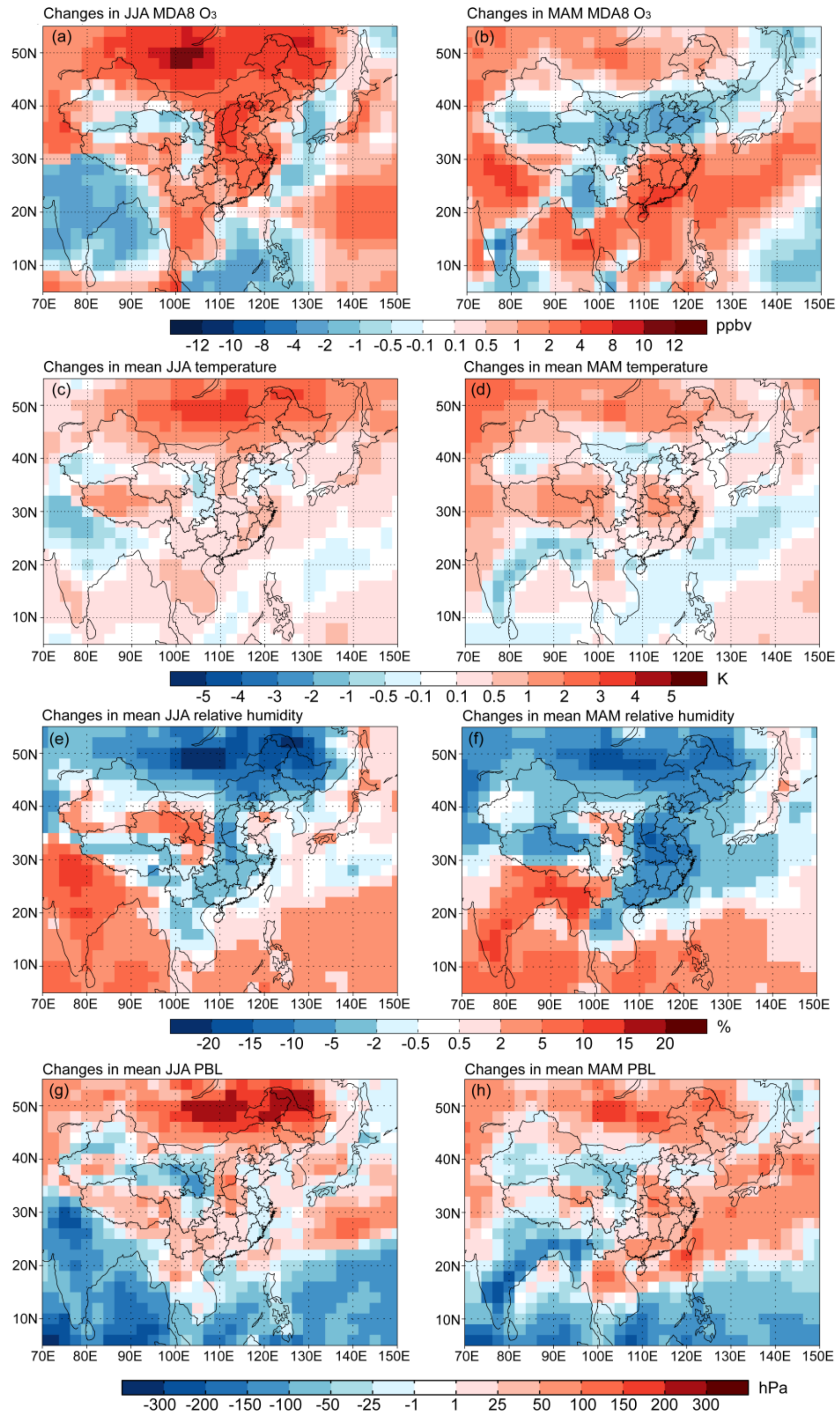


Figure 4. Changes in (a) surface maximum daily 8 h average ozone concentration (MDA8 $O_3$) in summer (JJA); (b) surface MDA8 $O_3$ in spring (MAM); (c) mean JJA temperature; (d) mean MAM temperature; (e) mean JJA relative humidity; (f) mean MAM relative humidity; (g) mean JJA planetary boundary layer (PBL); and (h) mean MAM PBL driven by changes in climate alone ([CTRL]–[SIM_CLIM]). Values are differences between the five-year averages over the present-day and historical periods.

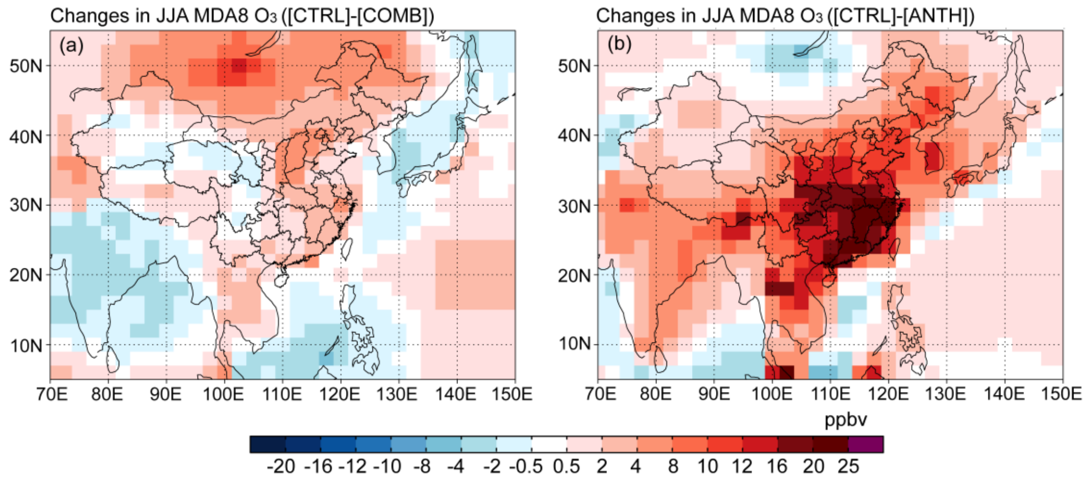


Figure 5. Changes in summertime (JJA) surface maximum daily 8 h average ozone concentration (MDA8 $O_3$) driven by changes in (a) climate, land cover and land use combined ([CTRL]–[SIM_COMB]); and (b) anthropogenic emissions alone ([CTRL]–[SIM_ANTH]).

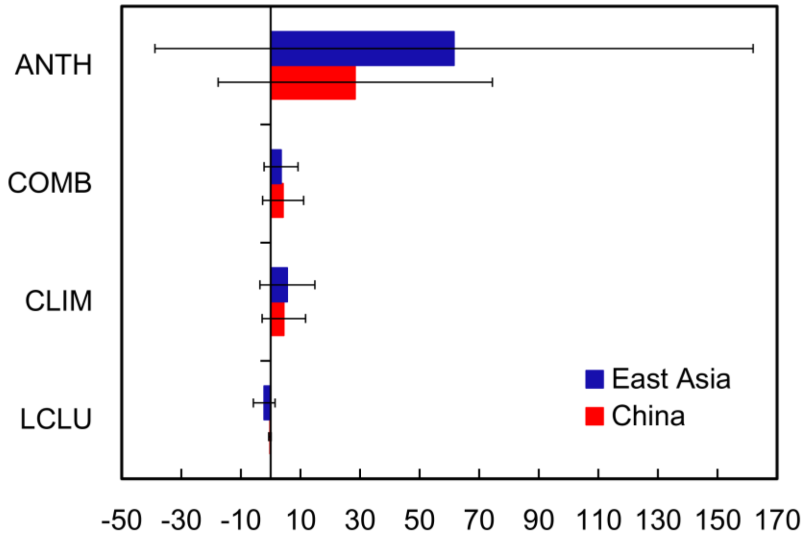


Figure 6. Estimates of ozone-related respiratory mortality (in 1000 deaths $yr^{-1}$) attributable to historical (1980–2010) changes in land cover and land use (LCLU), climate (CLIM), climate and LCLU combined (COMB), and anthropogenic emissions (ANTH) in all of East Asia and China. Uncertainty for each case represents the 95% confidence interval of the concentration response function.