



*Supplement of*

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

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## 1 **Supplementary Materials**

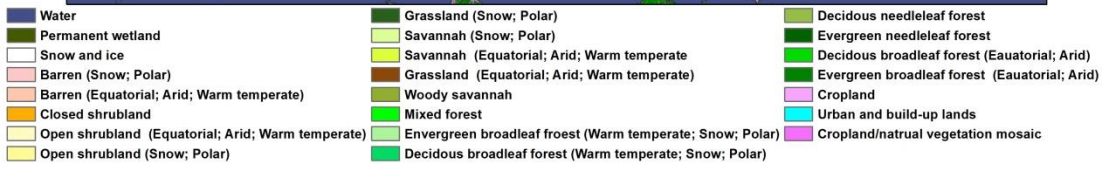
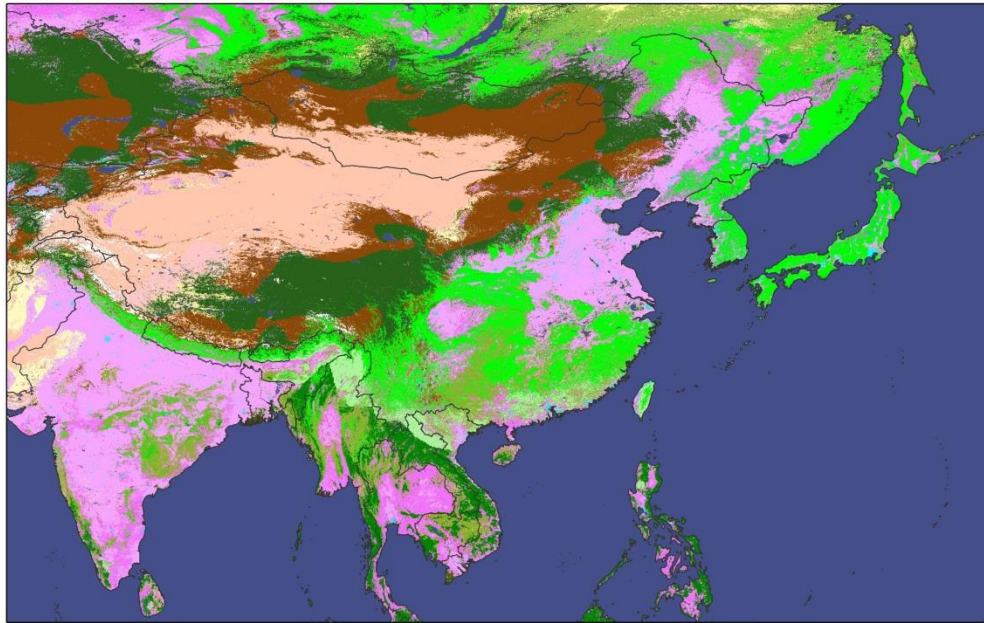
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### 3 *S1. Detail land cover and land use data*

4 The Moderate Resolution Imaging Spectroradiometer (MODIS) satellite-derived  
5 high-resolution land cover product for year 2010 is used as the baseline case in this work  
6 (MCD12Q1) ([https://lpdaac.usgs.gov/products/modis\\_products\\_table/mcd12q1](https://lpdaac.usgs.gov/products/modis_products_table/mcd12q1)). This  
7 product includes five land cover classification schemes. The MODIS land cover product  
8 includes five land cover classification schemes. Here we choose the classification scheme  
9 of IGBP (International Geosphere-Biosphere Program) with 17 land cover types,  
10 including 13 vegetation classes (evergreen needleleaf forest, evergreen broadleaf forest,  
11 deciduous needleleaf forest, deciduous broadleaf forest, mixed forest, closed shrublands,  
12 open shrublands, woody savannas, savannas, grasslands, permanent wetlands, croplands,  
13 cropland/natural vegetation mosaic) and 4 non-vegetated land types (water, urban and  
14 built-up, snow and ice, barren or sparsely vegetated). We further combine them with the  
15 Koppen main climate classes following Steinkamp and Lawrence (2011). The  
16 distribution of land cover and land use types used in this study are shown in Fig. S1.

17 To derived the land cover and land use in year 1980, we produce the potential land  
18 cover map by integrating multiple sources of data and information, including the China  
19 National land cover dataset (NLCD) for in the late 1980s and year 2005, MODIS-IGBP  
20 land cover types in 2005, Koppen main climate classes map, as well as and the  
21 harmonized historical cropland and urban land use dataset from the historical land cover  
22 projects of Representative Concentration Pathways (RCPs) for the period 1980-2005  
23 from Hurtt et al.(2011). The reconstructed cropland and urban areas are based on the  
24 HYDE model which combines numerous historical statistics, census data and  
25 satellite-derived current land cover from DISCover 2 data (Loveland et al., 2000) and  
26 Global Land Cover (GLC 2000) (Bartholome et al., 2002). The method we use to  
27 reconstruct the land cover and land use in 1980 is similar to that of Liu and Tian (2010),  
28 and is based on the MODIS-IGBP-Koppen LCLU in year 2005 as base year and applies  
29 appropriate calibration ratios to scale up/down the 2005 data. For instance, cropland and  
30 urban fractional coverage in year 1980 in each model grid cell is obtained by scaling  
31 up/down the MODIS-IGBP-Koppen value in 2005 with a calibration ratio derived from  
32 the slope of time-series linear regression of 1980-2005 harmonized RCP data. For each of  
33 the other natural vegetation types, the calibration ratio is an overall ratio derived from the  
34 slope of reduced major-axis regression between NLCD 1980s and NLCD 2005 available  
35 data over the whole spatial domain. It should be noted that the sum of fractional  
36 coverages of all PFTs including bareland of each grid cell is always constrained to unity.

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38

39 Fig. S1. Land cover and land use distribution in year 2010.

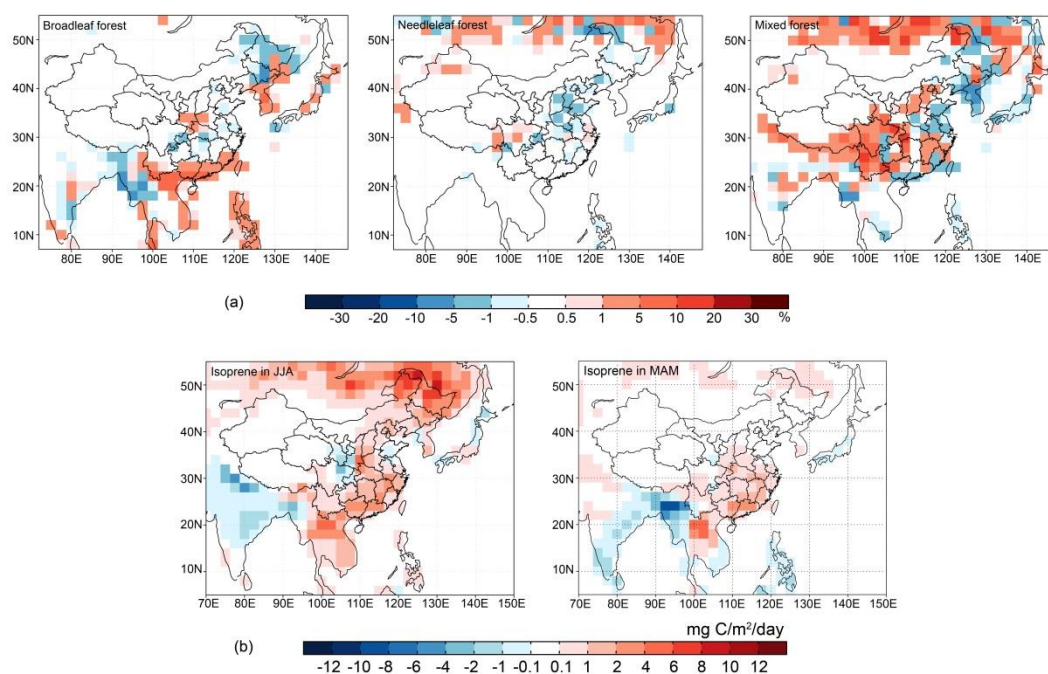
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## 41 *S2. Changes in biogenic VOC emissions*

42 Annual isoprene emission in East Asia decreases by 3% but increases by 2% in  
43 China specifically as a result of LCLU change alone over 1980-2010  
44 ( $[CTRL]-[SIM\_LCLU]$ ) (Table S4). Our calculated decreases (increases) in isoprene  
45 emission in East Asia (China) generally agree with Stavrakou et al. (2014). Seasonal and  
46 regional isoprene emission exhibits more pronounced changes in response to LCLU  
47 change. In summer (JJA), we find that the seasonal mean isoprene emission increases  
48 5-30% in central and southeastern China but decreases 5-20% over northeastern China,  
49 Korea and Myanmar (Fig.2b). Changes in JJA isoprene emission are consistent in spatial  
50 distribution with the changes in forest coverage (Fig. S2a). Increases in JJA isoprene  
51 emission are likely caused by the enhanced fractional coverage of broadleaf and mixed  
52 forests, while the reduction of isoprene emission results from the reduced coverage of  
53 both needleleaf and broadleaf forests over those regions. In spring (MAM), LCLU  
54 change alone reduces isoprene emission by as much as 40% in eastern China, Korea, and  
55 Southeast Asia (Fig. 3b). Such a decrease is primarily driven by reduced LAI in most of  
56 East Asia (Fig. 1b).

57 Climate change alone increases annual isoprene emission in East Asia by 7.4%  
58 ( $[CTRL]-[SIM\_CLIM]$ ) (Table S4). In summer, isoprene emission increases by 5-20% in  
59 most places of East Asia (Fig. S2b). In spring, isoprene emission increases by 5-40% in  
60 southern China and parts of Southeast Asia, while isoprene emission in Myanmar,  
61 northern China and Japan decreases by 5-30% due to climate change alone.

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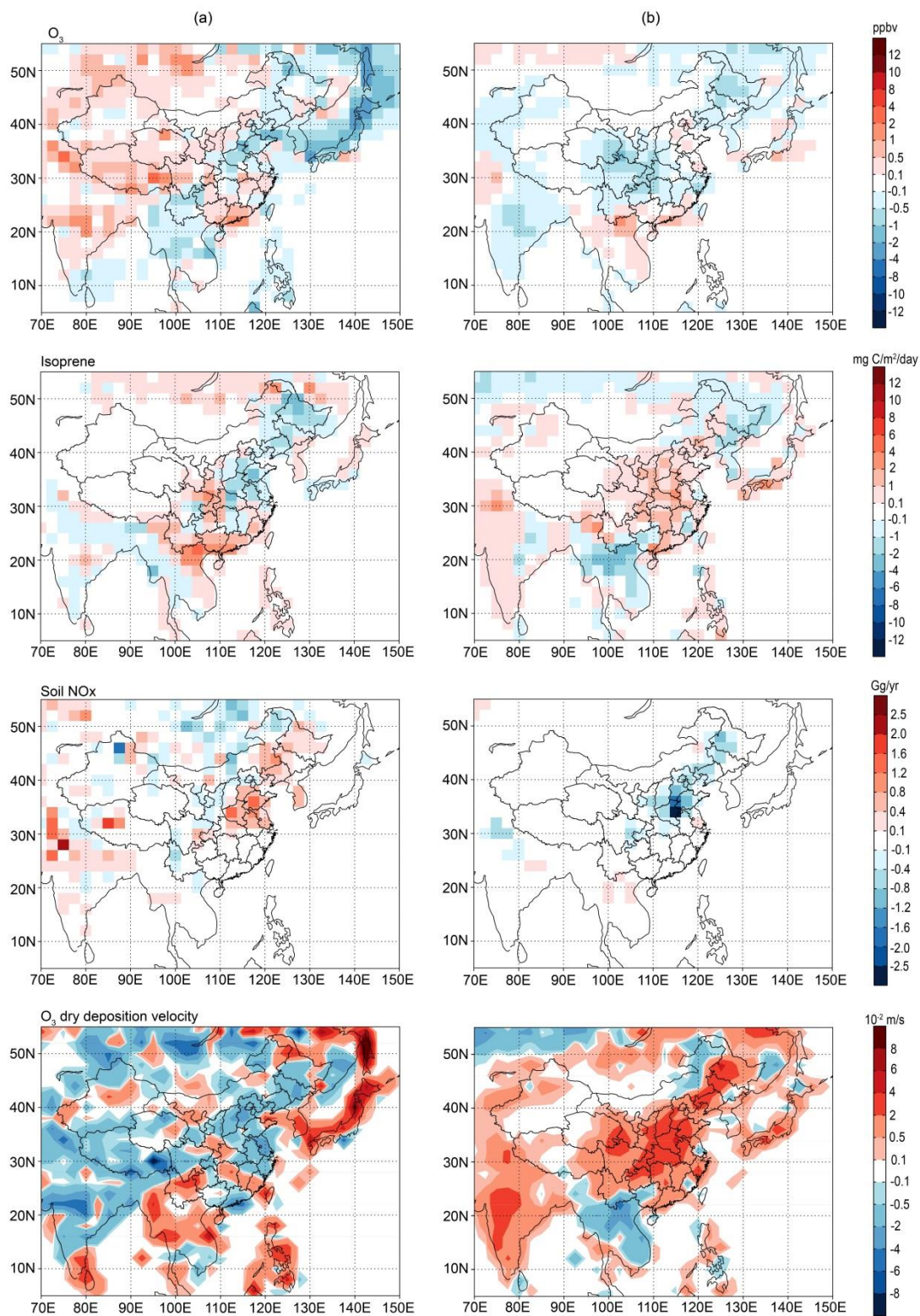


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64 Fig. S2. Changes in (a) fractional coverage of broadleaf, needleleaf, and mixed forests;  
65 and (b) in isoprene emission in summer (JJA) and spring (MAM) driven by climate  
66 change alone over 1980-2010.

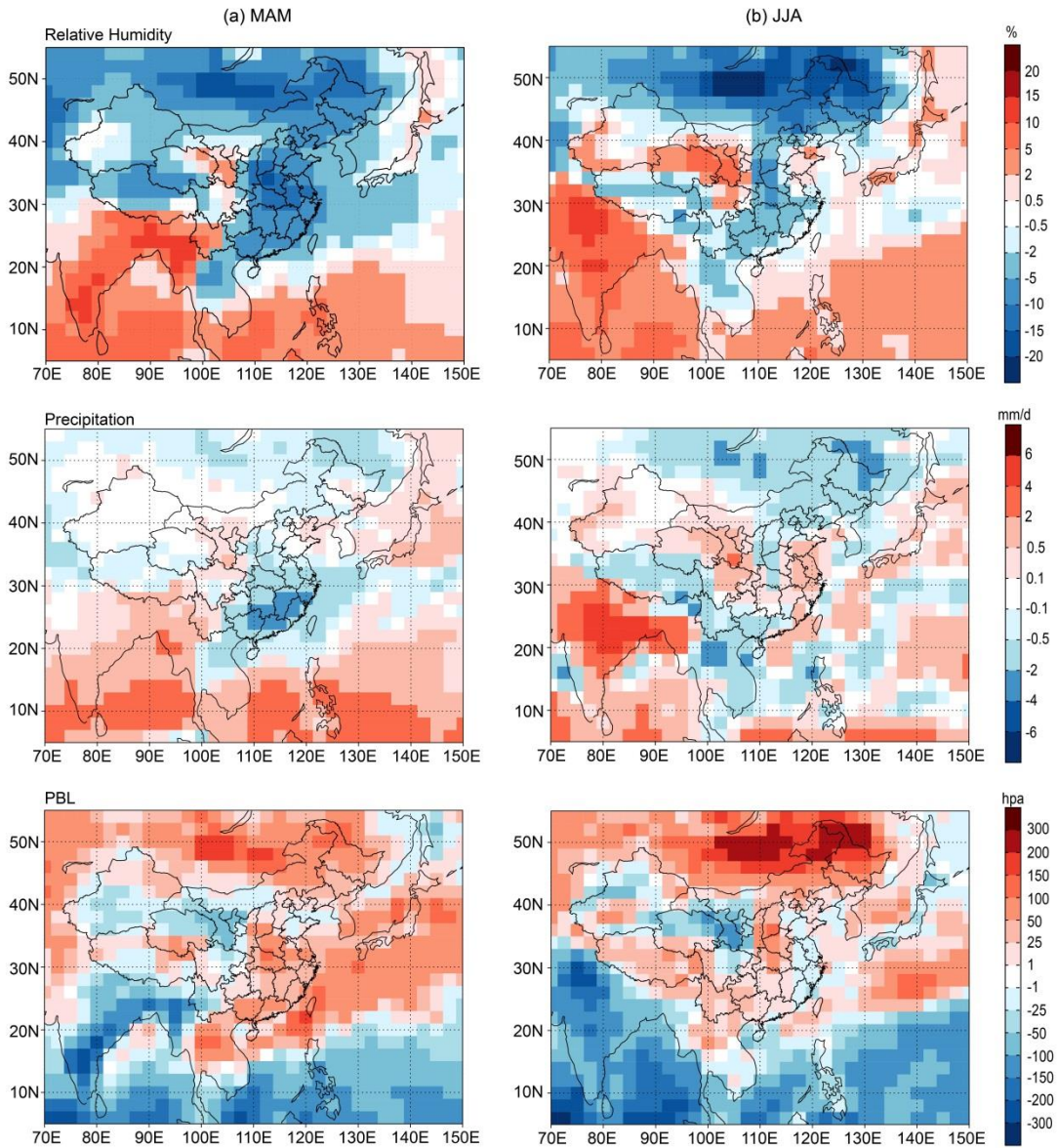
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68 **S3. Effects of 1980-2010 changes in vegetation distribution alone and in density alone**



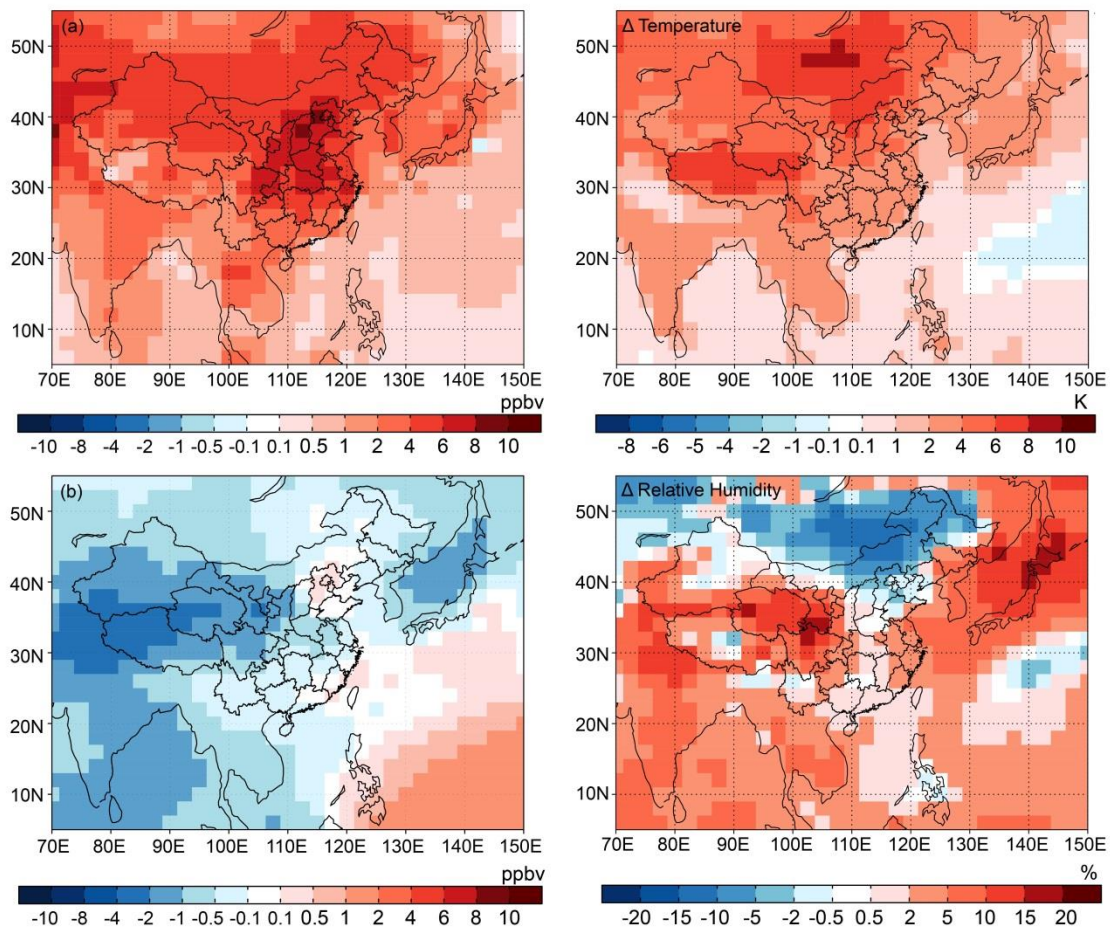
69  
 70 Fig. S3. Changes in summertime surface ozone, isoprene emission, soil  $NO_x$  emission,  
 71 and dry deposition velocity arising from changes in (a) vegetation distribution  
 72 (represented by PFT fractional coverage) alone ( $[CTRL\_2010]-[SIM\_PFT]$ ) and (b)  
 73 vegetation density (represented by LAI) alone ( $[CTRL\_2010]-[SIM\_LAI]$ ).  
 74

75 *S4. 1980-2010 changes in meteorological variables derived from MERRA*  
 76 *meteorological fields*



77  
 78 Fig. S4. Changes in (a) spring and (b) summer average relative humidity (RH),  
 79 precipitation, and planetary boundary layer (PBL) from the period 1981-1985 to the  
 80 period 2007-2011.  
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82 **S5. Effects of changes in temperature alone and relative humidity alone**



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Fig. S5. Changes in surface maximum daily 8-hour average ozone concentration (MDA8 O<sub>3</sub>) driven by changes in (a) temperature alone ([CTRL\_2010]-[SIM\_TMP]); and (b) relative humidity alone ([CTRL\_2010]-[SIM\_RH]).

88 ***S6. Method for assessing the health impact due to ozone air quality***

89 Since there are very limited studies reporting long-term ozone-related mortality in  
90 East Asia, we apply epidemiological concentration-response functions (CRFs) from  
91 American Cancer Society (ACS) in this study. As in Anenberg et al (2010) and Raquel et  
92 al (2013), we use surface ozone concentrations to estimate excess ozone-related  
93 respiratory mortality ( $\Delta Mort$ , unit: 1000 deaths per year per square km) by applying the  
94 following CRFs (Anenberg et al., 2010):

95 
$$\Delta Mort = y_0(1 - e^{-\beta\Delta X})Pop$$

96 where  $y_0$  represents the baseline mortality rate (unit: deaths per thousand people per year),  
97  $\beta$  is a concentration-response factor,  $\Delta X$  represents the differences in ozone  
98 concentrations (April-September 6-month averaged of 1-h daily maximum ozone  
99 concentration (Jerrett et al., 2009)), and  $Pop$  is the exposed population (unit: persons per  
100 square km). Consistent with ACS, we only assess ozone-related health impact for all  
101 adults aged 30 and above. We use gridded population of the world at approximately  
102  $0.5^\circ \times 0.5^\circ$  resolution from Socioeconomic Data and Applications Center (SEDAC),  
103 Columbia University (version 3 for future estimates in year 2010;  
104 <http://sedac.ciesin.columbia.edu/data/set/gpw-v3-population-density-future-estimates>,  
105 accessed by 7 November 2014) and United Nations estimates of 2010 population by  
106 age-group per country (World Population Prospects, 2012 revision,  
107 <http://esa.un.org/unpd/wpp/Excel-Data/population.htm> , accessed by 7 November 2014)  
108 to calculate the fraction of population aged 30 and above at the country level and the  
109 exposed population aged 30 and above in East Asia. Then, the exposed population over  
110 30 is regridded to  $2^\circ \times 2.5^\circ$  resolution consistent with that of the simulated ozone  
111 concentrations.

112 Baseline mortality rates for respiratory disease are calculated from the World Health  
113 Organization 2000-2012 country-level cause-specific mortality for population aged 30 or  
114 above ([http://www.who.int/healthinfo/global\\_burden\\_disease/estimates/en/index1.html](http://www.who.int/healthinfo/global_burden_disease/estimates/en/index1.html),  
115 accessed by 7 November 2014). The concentration-response factor is derived from  
116 relative risks (RR) estimated in long-term epidemiological studies, which is represented  
117 as a log-linear relationship between ozone concentration and RR ( $\beta = \ln(RR)/10$ ) (Jerrett  
118 et al., 2009).

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Table S1. PFT-specific emission factors\* for biogenic VOC emissions used in this study (units:  $\mu\text{g m}^{-2} \text{h}^{-1}$ ).

PFTs	Compound Class	Isoprene	Monoterpenes						MBO	Acetone	
			$\alpha$ -Pinene	$\beta$ -Pinene	Myrcene	Sabinene	Limonene	3-Carene			Ocimene
Envergreen broadleaf forest ( <i>warm temperate; snow; polar</i> )		10000	400	130	30	50	80	30	120	0.01	240
Deciduous broadleaf forest ( <i>warm temperate; snow; polar</i> )		10000	400	130	30	50	80	30	120	0.01	240
Deciduous needleleaf forest		1	510	200	60	40	130	80	60	0.01	240
Evergreen needleleaf forest		3000	500	300	70	70	100	160	70	60	240
Deciduous broadleaf forest ( <i>equatorial; arid</i> )		7000	600	120	80	80	80	40	150	0.01	240
Evergreen broadleaf forest ( <i>equatorial; arid</i> )		7000	600	120	80	80	80	40	150	0.01	240
Closed shrubland		4000	200	100	30	50	60	30	90	0.01	240
Open shrubland ( <i>equatorial; arid; warm temperate</i> )		2000	200	100	30	50	60	30	90	0.01	240
Open shrubland ( <i>snow; polar</i> )		2000	200	100	30	50	60	30	90	0.01	240
Grassland ( <i>snow; polar</i> )		1600	2	1.5	0.3	0.7	0.7	0.3	2	0.01	80
Savannah ( <i>snow; polar</i> )		1600	2	1.5	0.3	0.7	0.7	0.3	2	0.01	80
Savannah ( <i>equatorial; arid; warm temperate</i> )		800	2	1.5	0.3	0.7	0.7	0.3	2	0.01	80
Grassland ( <i>equatorial; arid; warm temperate</i> )		800	2	1.5	0.3	0.7	0.7	0.3	2	0.01	80
Woody savannah		4000	300	150	50	70	100	100	150	0.01	240
Cropland		1	2	1.5	0.3	0.7	0.7	0.3	2	0.01	80
Mixed forest	Assumed the coverage are equally divided into 4 types of broadleaf trees and 2 types of needleleaf trees										
Cropland/natural vegetation mosaic	Assumed the coverage are divided into cropland (40%), shrubland (30%) and grassland (30%)										

\* Guenther et al. (2012)

Table S2. Method for merging 23 land types into 5 MEGAN PFTs

No.	MODIS_Koppen land type	MEGAN PFT
1	Water	-
2	Permanent wetland	-
3	Snow and ice	-
4	Barren (snow; polar)	-
5	Unclassified	-
6	Barren (equatorial; arid; warm temperate)	-
7	Closed shrubland	Shrub
8	Open shrubland (equatorial; arid; warm temperate)	Shrub
9	Open shrubland (snow; polar)	Shrub
10	Grassland (snow; polar)	Grass
11	Savannah (snow; polar)	Grass
12	Savannah (equatorial; arid; warm temperate)	Grass
13	Grassland (equatorial; arid; warm temperate)	Grass
14	Woody savannah	Shrub
15	Mixed forest	Assume Mixed forest consist of 30% broadleaf trees and 70% needleleaf trees
16	Evergreen broadleaf forest (warm temperate; snow; polar)	Broadleaf trees
17	Deciduous broadleaf forest (warm temperate; snow; polar)	Broadleaf trees
18	Deciduous needleleaf forest	Needleleaf trees
19	Evergreen needleleaf forest	Needleleaf trees
20	Deciduous broadleaf forest (equatorial; arid)	Broadleaf trees
21	Evergreen broadleaf forest (equatorial; arid)	Broadleaf trees
22	Cropland	Crop
23	Urban and built-up lands	-
24	Cropland/natural vegetation mosaic	Assume this type composes of 40% crop, 30% shrub and 30% grass.

Table S3. Mapping 23 land types into Olson land types used for dry deposition

NO	MODIS_Koppen land types	Olson ID	Olson land types*
1	Water	0	water
2	Permanent wetland	45	wetland
3	Snow and ice	17	Ice
4	Barren (snow; polar)	53	Barren tundra
5	unclassified	-	Not used
6	Barren (equatorial; arid; warm temperate)	8	Desert
7	Closed shrubland	47	Shrub
8	Open shrubland (equatorial; arid; warm temperate)	47	Shrub
9	Open shrubland (snow; polar)	42	Shrub/grass (cold)
10	Grassland (snow; polar)	40	Shrub/grass (cool)
11	Savannah (snow; polar)	40	Shrub/grass (cool)
12	Savannah (equatorial; arid; warm temperate)	-	-
13	Grassland (equatorial; arid; warm temperate)	41	Shrub/grass (hot and mild)
14	Woody savannah	32	Dry tropical woods
15	Mixed forest	24	Mixed forest
16	Evergreen broadleaf forest (warm temperate; snow; polar)	25	Cool broadleaf forest
17	Deciduous broadleaf forest (warm temperate; snow; polar)	25	Cool broadleaf forest
18	Deciduous needleleaf forest	21	Conifer boreal forest
19	Evergreen needleleaf forest	22	Conifer
20	Deciduous broadleaf forest (equatorial; arid)	29	Tropical broadleaf
21	Evergreen broadleaf forest (equatorial; arid)	29	Tropical broadleaf
22	Cropland	31	Agricultural
23	Urban and build-up lands	1	Urban
24	Cropland/natural vegetation mosaic	57	Mixed wood/open

\* 72 Olson land types are further corresponding to 11 surface types for calculating dry deposition in GEOS-Chem model (Wesely, 1989; Olson, 1992; Jacob et al., 1990, 1992; Wang et al., 1998)

Table S4. Simulated changes in biogenic hydrocarbon emission and soil NO<sub>x</sub> emission due to LCLU change, climate change, and combined climate and LCLU change in East Asia. The domain of East Asia is 5.5 °-56.0 °N, 69.0 °-149.0 °E.

Species	CTRL	SIM_LCLU (changes, %)	SIM_CLIM (changes, %)	SIM_COMB (changes, %)
<b>Biogenic hydrocarbons (Tg C yr<sup>-1</sup>)</b>				
Isoprene	34.700	35.635(-2.6)	32.316 (+7.4)	33.271 (+4.3)
Monoterpenes	9.389	9.370 (+0.2)	8.859 (+6.0)	8.855 (+6.0)
Methyl Butenol	0.239	0.241 (-0.7)	0.216 (+11.0)	0.218 (+9.8)
Farnesene	0.240	0.252 (-4.8)	0.225 (+6.9)	0.239 (+0.7)
b-Caryophyllene	0.326	0.339 (-3.8)	0.304 (+7.5)	0.320 (+2.2)
Other sesquiterpenes	0.659	0.687 (-4.0)	0.614 (+7.4)	0.650 (+1.9)
Other monoterpenes	1.509	1.482 (+1.8)	1.413 (+6.8)	1.393 (+8.3)
Acetone	5.116	5.173 (-1.1)	4.920 (+4.0)	4.979 (+2.7)
PRPE (lumped >= C3 alkenes)	0.992	1.019 (-2.6)	0.924 (+7.4)	0.952 (+4.3)
Total	53.172	54.199 (-1.9)	49.790 (+6.8)	50.873 (+4.5)
<b>NO<sub>x</sub> (Tg N yr<sup>-1</sup>)</b>				
Soil NO <sub>x</sub>	2.067	2.053 (+0.7)	2.019 (+2.4)	2.000 (+3.3)
Fertilizer NO <sub>x</sub>	0.742	0.741 (+0.1)	0.771 (-3.9)	0.771 (-3.9)
<b>Ozone burden (Tg) (up to 2 km)</b>				
Annual mean in EA	5.638	5.642(-0.071)	5.609(+0.52)	5.615(+0.41)
Summer mean in EA	5.946	5.961(-0.25)	5.795(+2.61)	5.809(+2.36)

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