Sensitivity of China's ozone air quality to 2000-2050 global changes of climate and emissions

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Abstract: We use a global chemical transport model (GEOS-Chem) driven by the GISS GCM to investigate the effect on China's ozone air quality from 2000-2050 global changes in climate and anthropogenic emissions of ozone precursors. The climate and emission effect in combination will increase afternoon mean surface ozone over China by an annual average of 8.7 ppbv, of which 65% is attributed to the projected increases in global (excluding China) anthropogenic emissions, 37% to Chinese emission increases, and a small negative contribution from climate change (-1.8%) which reduces ozone lifetime. Afternoon mean surface ozone over the Tibet Plateau is projected to increase by 10-15 ppbv in summer, attributed to increasing emissions from neighboring countries in addition to China, suggesting the crucial need for an effective trans-boundary pollution control policy to protect the fragile ecosystems and glaciers over this region. Over Central East China (CEC), a region of large population and intensive agriculture, the 2000-2050 global changes increase annual afternoon mean surface ozone by 9.0 ppbv, of which the Chinese emission change makes the largest contribution (49%), followed by global emission change (43%) and climate change (+7.9%). The change in Chinese anthropogenic emissions of nitrogen oxides (NOx) alone is responsible for 80% of the overall surface ozone increase resulting from the Chinese emission change of all ozone precursors, indicating that aggressive control of anthropogenic NO_x emissions is the first priority for Chinese policy-makers to mitigate ozone pollution problems in the future. The climate change penalty projected over CEC is attributed to increasing biogenic emissions of volatile organic compounds (VOCs) as well as to changing meteorological factors such as reduced planetary boundary layers.

We find that 2000-2050 climate change will decrease the sensitivity of surface ozone to Chinese anthropogenic emissions over West China due to the accelerated ozone destruction rate and reduced transport from CEC, but increase this sensitivity over CEC by 10% as a result of the coupling between anthropogenic NO_x and biogenic VOCs. This implies that the emission controls over China need to be more aggressive in the future, with the first priority being control of anthropogenic NO_x emissions.

1. Introduction

Rapid changes of the global climate system predicted for the next 50-100 years [IPCC,2001, 2007] will have important implication for air quality. Direct consequences of climate change for air quality may result from changes in temperature, precipitation, humidity, cloud cover, cyclone frequency, boundary layer mixing, and wet convective ventilation [McCabe et al., 2001; Meleux et al. 2007; Held and Soden, 2000; Forkel and Knoche 2006; Murazaki and Hess,2006;Lambert and Fyfe, 2006; Hogrefe et al., 2004; Rind et al., 2001; Wu et al., 2008a; Johnson,et al.,1999]. Natural emissions of VOCs from vegetation, NO_x from soil and lightning, and dust are all strongly dependent on climatic factors and are therefore sensitive to climate change. The frequency of forest fires is expected to increase in the 2050 climate, resulting in an adverse effect on air quality [Spracklen et al.， 2009].

Air quality depends on both emissions and weather. A chemical transport model (CTM) driven by future climate archived from a general circulation model (GCM) is typically used to capture the complex coupling between climate change and parallel changes in emissions [Grewe et al., 1999, 2001; Johnson et al., 1999, 2001; Zeng and Pyle, 2003; Grenfell et al., 2003; Allison et al., 2006; Wu et al., 2008a, 2008b; Jacob and Winner, 2009 and references therein]. During the past decades, GCM-CTM studies on regional air quality have been mostly concerned with developed regions, U.S. and Western Europe in particular, focusing on the climate change penalty which tends to offset the benefit of expensive domestic emission reductions [Jacob and Winner, 2009]. A number of studies suggested that climate change alone would decrease global mean surface ozone in the future, largely driven by increased humidity [Wu et al., 2008b; Lin et al., 2008; Nolte et al., 2008; Brasseur et al., 2006; Grewe et al., 1999, 2001; Johnson et al., 1999, 2001; Liao et al.,2006; Unger et al., 2006]. Dentener et al.[2006], in a survey of results from 10 global GCM-CTM models, suggested that annual mean surface ozone in the North Hemisphere will decrease 0.8 ppb as a result of 2000-2030 climate change. Climate change will likely enhance the stratosphere-troposphere exchange[Zeng and Pyle, 2003; Hauglustaine et al.,2005]. Johnson et al. [1999] indicated that when both climate and emission changes were taken into account, the response of global tropospheric ozone during the period 1990-2075 will be an increase of 6.4 ppb. Wu et al. [2008b] projected that the 2000-2050 emission changes will lead to a 17% increase of tropospheric ozone burden globally, while climate change will contribute to an additional increase of 1.6%.

For large developing nations such as China, the challenge for air quality management still relies heavily on controlling rapid increasing domestic emissions. Chinese anthropogenic emissions of NO_x have increased by 70% from 1995 to 2004 [Zhang et al., 2007]. The A1B socioeconomic scenario for the 21st century developed by the IPCC [IPCC,2007] projects increasing emissions of ozone precursors in developing countries including China, in contrast to decreasing emissions in OECD countries. Economic development in developing countries is the driving factor for the overall global increase of emissions for 2000-2050. Climate change adds another dimension of complexity in predicting the response of increasing anthropogenic emissions on air quality. However, few studies have examined the consequences of parallel changes in climate and emissions on regional air quality in developing countries, despite the increasing importance of rising emissions from them on global atmospheric environment.

The present study uses a global chemical transport model (GEOS-Chem) driven by the GISS GCM to investigate the effect on China's ozone air quality from 2000-2050 global changes in climate and anthropogenic emissions of ozone precursors. Global anthropogenic emissions in 2050 are adopted from the IPCC A1B scenario which projects rapid global economic growth along with the introduction of more energy-efficient technologies, reductions in regional differences of per capita income, and balanced energy generation from fossil and alternative fuels. Natural emissions of ozone precursors are modeled as a function of climatic factors. For implications on emission control policies, this study decomposes 2000-2050 global changes into independent changes in three factors: climate, Chinese anthropogenic emissions (CHE) and anthropogenic emissions from the rest of the world (RWE). Through an ensemble of model sensitivity analysis, we evaluate the separate and combined effects on surface ozone from the projected changes in the three factors. The implication of our analysis for air quality policies in China will be discussed.

We begin in Section 2 to introduce the GCM-CTM model used in this study and summarize the sensitivity simulations. The climate-only, CHE-only, and RWE-only effect on surface ozone over China is discussed separately in Section 3 and 4. The impact of climate on the CHE-only and RWE-only effect will be discussed in Section 5. Section 6 will present the combined effects of parallel changes in both climate and emissions and compare the relative contributions from the individual factors. Concluding remarks are given in Section 7.

2. Model and Simulations

2.1. Model description

We use the NASA/GISS GCM 3 [Rind et al., 2007] to simulate the present-day and 2050 climate based on the IPCC A1B emission scenario of greenhouse gases. Meteorological output from the GISS model was archived to drive the GEOS-Chem global chemical transport model [Wu et al.,2008a, 2008b]. The GISS model version used here has a horizontal resolution of 4^o (latitude) x 5^o (longitude) and 23 sigma levels up to 0.002 hPa. The GEOS-Chem model includes detailed mechanism to simulate the ozone-NOx-VOC-aerosol chemistry in the troposphere. The model setup is the same as in Wu et al. [2008a] and we do not give detailed description here.

The 2000-2050 changes in global anthropogenic emissions of ozone precursors are based on the IPCC A1B emission scenario. We applied the same scaling factor as in *Wu et al.* [2008a] to obtain 2050 emissions on the basis of 2000 emissions. This results in the same 2000-2050 emission trends by world regions as in *Wu et al.* [2008a] but slightly different global total emissions because of some updates in the present-day emission inventories. Biogenic emissions of nonmethane VOCs (NMVOCs) are adopted from the Model of Emissions of Gases and Aerosols from Nature (MEGAN) inventory [*Guenther et al.*, 2006], in which the present-day emissions of isoprene over China is 50% smaller than those from the GEIA inventory used by *Wu et al.* [2008a]. Biomass burning emissions are taken from the GFED-2 inventory [*van der Werf et al., 2006*].Although rising levels of atmosphric CO_2 may affect the natural emissions of isoprene, monoterpenes and other VOCs [Sanderson et al., 2003; Rosenstiel et al., 2003], we did not take this process into account. The methane mixing ratios used for the present-day scenario is specified with a global mean of 1750 ppb and 5% inter-hemispheric gradient, which will rise to 2400 ppb by 2050 with no hemispheric gradient in the future scenario.

This study focuses on China where rapid economical growth is projected. Table 1 summarizes the changes in anthropogenic and biogenic emissions for the world and for China, as projected by the A1B scenario. During the 50 years, anthropogenic NO_x emissions are projected to increase by 79% globally and by 116% in China. Anthropogenic emissions of NMVOCs will increase by 160% globally and by 102% in China. In contrast, for example, a decrease of 40% in fossil fuel NO_x emissions is projected in the United States due to effective emission controls assumed for developed countries. Although anthropogenic emissions of CO will increase by 20% globally, they will decrease by 10% in China in the future. We do not account for the change in stratosphere-troposphere ozone exchange (STE) in the simulation, which is kept constant as a mean STE ozone flux of 500 Tg/yr. The possible coupling effect of climate change on biomass burning is not considered in this study. We applied the same species-specific scaling factor as in *Wu et al.* [2008a] to obtain the biomass burning emissions in 2050 on the basis of 2000 emissions.

The model simulations were conducted for eight cases described below in order to decompose the contributions from climate and emission changes. To account for the interannual variability, each simulation was preformed for three years, 1999-2001 for the present-day climate and 2049-2051 for the future climate.

2.2. Sensitivity simulations

Following the typical GCM-CTM approach, we separate the contributions from climate and emission changes through a sensitivity analysis. To cover the full array of combinations for 2000-2050 changes in climate, Chinese anthropogenic emissions (CHE), and rest of the world anthropogenic emissions (RWE), we set up eight cases summarized schematically in Fig 1. The X-axis denotes the 2000-2050 changes in CHE, Y-axis RWE, and Z-axis climate change. The case pairs on each axis can be used to analyze the effect of the single factor associated with that axis on ozone. The difference between the case pairs along X, Y, and Z axes is referred to as the CHE-only effect, RWE-only effect, and climate-only effect on surface ozone, respectively. For example, the difference between case A and B (B-A) indicates the CHE-only effect on surface ozone under the present-day climate. Case F-E indicates also the CHE-only effect, but under the 2050 climate.

As our study focuses on ozone air quality, mean afternoon ozone mixing ratio at the surface is the metric adopted to evaluate the influence of each case on ozone air quality over China. The numbers displayed along the axes in Fig 1 represent the differences in the annual-mean value of this metric between the cases. Along each axis, the first number represents the difference averaged over China and the second (in parenthesis) the difference averaged over Central Eastern China (rectangular region in Fig 2a), a region of large population and intensive agriculture. Central eastern China (CEC) accounts for 68% of China's population, 76% of its GDP, and 62% its anthropogenic NO_x emissions in present days. Unless noted otherwise, all the results reported in this paper refer to mean afternoon mixing ratios at the surface.

3. Effect of climate change alone

The 2000-2050 climate change will result in an increase of 1.8K in annual-mean surface air temperature and 5% increase in annual precipitation in China. Specific humidity in surface air will increase by 12% in China. The 2000-2050 changes in summertime precipitation, PBL height, and convective mass flux simulated by the GISS GCM are presented in Fig 3a-c respectively. There is a maximal decrease of 20% in PBL height and a slight decrease in convective mass flux over CEC. This suggests that the 2000-2050 climate change will reduce ventilation rates in the boundary layer over

CEC. We calculate that the 2000-2050 climate change will result in a 42% increase in isoprene emissions in China as a result of higher temperature and solar radiation [Guenther et al., 1995; Wang et al., 1998]. The increase is largest over South China in summer, largely determined by the large temperature increase and high forest coverage over this region. Higher soil temperature and moisture contribute to increasing soil NO_x emissions in the 2050 climate [Yienger and Levy,1995; Wang et al., 1998]. The lighting flash frequency will increase in the 2050 climate due to deeper convection [Price and Rind, 1992; Wang et al., 1998;Li et al., 2005], resulting in a 17% increase in lighting-NO_x emissions globally.

The distribution of annual-mean surface ozone simulated for the present-day climate and emissions (Case A) is illustrated in Fig 2a. The Case A simulation provides a benchmark to compare with the results from other cases. The ozone differences between Case E and Case A, representing the climate-only effect on surface ozone under the present-day anthropogenic emissions, are displayed in Fig 2b (annual-mean) and 2c (summer-mean). The annual-mean effect averaged over China is a decrease of 0.21 ppbv in surface ozone. The decrease is larger (0.5-1 ppbv) over northwest and northeast China, whereas an average increase of 0.56 ppb is found over CEC. The climate-driven ozone increase over CEC is most pronounced and spatially extensive in summer, reaching to a regional-mean of 1.7 ppbv and a maximum of 5 ppbv in some areas (Fig 2c). The maximal ozone increase over CEC corresponds well with the maximal decrease in PBL height and a slight decrease in convective mass flux shown in Fig 3. This suggests that the reduced ventilation rates in the boundary layer over CEC will contribute to surface ozone increases over this region. In contrast, the ozone decrease over northwest China is associated with increases in convection and precipitation. Previous studies have suggested similar spatial heterogeneity of the climate-only effect on surface ozone, with increasing ozone in polluted continental regions while decreases in relatively clean areas [Brasseur et al., 1998; Grewe et al.2001; Johnson et al., 1999, 2001; Collins et al., 2003; Murazaki and Hess, 2006;Wu et al.,2008a].

The following reactions (R1-R7) present the main reactions of O_3 formation and destruction in the troposphere [Murazaki and Hess, 2006]. The increase of water vapor in the 2050 climate accelerates the destruction of ozone through R2, which results in decreased ozone lifetime and therefore lower surface ozone over remote regions, particularly over the oceans [Wu et al., 2008b; Lin et al., 2008a; Nolte et al., 2008]. Climate change will also reduce PAN stability due to higher temperature, leading also to reduced ozone background over continental regions [Johnson et al., 1999; Murazaki and Hess,2006]. Climate change will also lead to increasing HO_2, which plays an important role in destructing O_3 in low-NOx regions through R4 [Lelieveld et al., 2002; 2004]. However, over regions where NO_x is in abundant supply, increases in HO_2 will lead to enhanced ozone production through R5-R7 [Murazaki and Hess,2006].

$$R1 \quad O_3 + hv \rightarrow O(^1D) + O_2$$
$$R2 \quad O(^1D) + H_2O \rightarrow 2OH$$
$$R3 \quad OH + CO(+O_2 + M) \rightarrow HO_2 + CO_2(+M)$$
$$R4 \quad HO_2 + O_3 \rightarrow OH + 2O_2$$
$$R5 \quad HO_2 + NO \rightarrow NO_2 + OH$$
$$R6 \quad NO_2 + hv \rightarrow NO + O$$
$$R7 \quad O + O_2 + M \rightarrow O_3 + M$$

The decrease of surface ozone over northeast and northwest China can be attributed to reduced global background ozone in the future climate, as background ozone is the dominant component of surface ozone over this region where anthropogenic emissions are small [Wang et al., 2010b]. The decrease represents a ‘climate change benefit’ as a result of the 2000-2050 climate change. In contrast, increasing surface ozone over CEC represents a ‘climate change penalty’ [*Wu et al.*, 2008a], because it implies that additional emission controls will be required to meet a given ozone air quality target over this populated region in the future climate. Under the present-day emissions, the climate change penalty over CEC shows an annual average of 0.56 ppbv and a summer-average of 1.71 ppbv. Given the large population in CEC (currently 874 million in 2005), this climate change penalty has significant implications for public health. In this region, the climate change penalty arises from the combination of enhanced natural emissions of ozone precursors (c.f. Table 1) and changes in meteorological factors including higher temperature, higher water vapor content, and reduced PBL as discussed above. To compare the relative importance between biogenic

VOCs and meteorology on ozone, we conducted a sensitivity simulation in which biogenic emissions of VOCs over China in the 2050 climate were scaled down to emission levels in 2000. In this case, the annual climate change penalty over CEC is reduced to 0.32 ppbv. This implies that 43% of the climate change penalty could be attributed to increases in biogenic emissions of VOCs, with the balance resulting from changes in meteorology.

4. Effect of emission changes alone

4.1. CHE changes alone

The difference between Case B and Case A suggests that the 2000-2050 change in CHE alone will increase annual mean surface ozone by an average of 3.2 ppbv for all of China under the present-day climate and RWE. The CHE-only effect on surface ozone distribution is illustrated in Fig 4a-c for the annual, summer, and spring conditions respectively. The summer and spring conditions are of particular interest because the ozone increase is expected to be largest in summer while Asian outflow to the Pacific typically peaks in spring [Liu et al., 2002]. The largest increase of annual-mean surface ozone is in the southeast and southwest, reaching 5-10 ppbv. The CHE-driven surface ozone increase is more than 2 ppbv all over China, with a maximum increase of 9.0 ppbv over CEC in summer. Over the southern regions (south of 30^{o}N), the 2000-2050 change in CHE will result in increasing surface ozone in all seasons. In contrast, no significant increase is found north of 35^{o}N in winter and spring because lower temperature and weak solar radiation over this region are the limiting factors for ozone formation instead of precursor emissions.

We conducted a sensitivity analysis in which we considered only the 2000-2050 changes in anthropogenic NO_x emissions in China. Under the 2000 climate, the projected increase in Chinese NO_x emissions alone is responsible for 86% and 80% of the overall increases in surface ozone over China and CEC, respectively. This indicates that aggressive control of anthropogenic NO_x emissions is the first priority for Chinese policy-makers to mitigate ozone pollution problems in the future, especially in the populated eastern region.

We find that the 2000-2050 change in CHE will increase annual mean global surface ozone by 0.4 ppbv. According to the A1B emission scenario, China contributes 20% and 10% of the 2000-2050 global increase of anthropogenic emissions of NO_x and VOCs respectively. Although the coarse resolution (4^o x 5^o) of the model makes it impossible to actually resolve emission changes across the country boundaries, increasing CHE seems to has a large impact on surface ozone over north India. In spring, the season of maximum pollution export from East Asia to the Pacific [Liu et al., 2002], the 2 ppbv-difference isopleth of ozone at the surface extends eastward from China's coasts up to 150^oE in central Pacific (Fig 4c), while the 2 ppbv-difference isopleth in the summer is confined to west of 130^oE (Fig 4b).

4.2. RWE changes alone

The difference between Case C and Case A suggests that the 2000-2050 change in RWE alone will increase annual mean surface ozone by an average of 5.7 ppbv and 3.7 ppbv for all of China under the present-day climate and CHE. The distribution of surface ozone change is illustrated in Fig 5a-b for annual and summer conditions respectively. A general feature is that the effect of RWE change alone shows a decreasing gradient from west to east and from south to north, reflecting different emission trends in surrounding countries/regions. In the present day, anthropogenic emissions of ozone precursors from Europe and North America have significant impact on ozone background over Northeast China, particularly in the springtime [*Wild et al.*, 2004; *Wang et al.*, 2010b]. Although emissions from Europe and U.S. are projected to decrease in the future, no significant change on surface ozone is found over Northeast China all year around because of the compensating effect of increasing emissions from developing countries. The simulated ozone increase along China's east coast is largest in summer when the prevailing monsoonal wind is southeasterly, bringing enhanced ozone exported from Southeast Asia where anthropogenic emissions will increase rapidly from 2000 to 2050. The largest surface ozone increase is in southwest China and the Tibet Plateau, reaching an annual mean increase of 6 ppbv and 15 ppbv in summer respectively. This can be largely attributed to increasing anthropogenic emissions from India. The A1B scenario forecasts that Indian anthropogenic emissions of NO_x and VOCs will increase by 800% and 300% respectively from 2000 to 2050. Over the pristine Tibetan Plateau region, the impact of the RWE change on surface ozone is 100% larger than that of the CHE change all year around. The challenge for policy-makers to protect the fragile ecosystems over the Tibetan Plateau and Himalayan Glaciers is to find an effective trans-boundary pollution control policy involving both China and India.

5. Effect of climate change on ozone sensitivity to emissions

The large number of cases constructed in this study make it possible to study the effect of climate change on ozone sensitivity to domestic and foreign emissions. Here we investigate whether a given change in emissions (domestic or foreign) will result in the same magnitude of ozone change in the future climate as compared with the present day. The discussion in this section is different from the climate-only effect or emission-only effect discussed above in that it focuses on the difference in the emission-only effect between the present-day and future climate. For example, the difference between Case A and Case B (B-A) yields the CHE-only effect on surface ozone under the present-day climate. Case F-E also yields the CHE-only effect, but under the 2050 climate. The difference between the two CHE-only effects represents the climate-driven change in the sensitivity of ozone to domestic emissions.

5.1. Sensitivity of CHE-only effect to climate

As summarized in Fig 1, the CHE-only effect on surface ozone over China is an annual increase of +3.2 ppbv under the 2000 climate (Case B-A), increasing by 6% to +3.4 ppbv under the 2050 climate (Case E-F). Over CEC, the CHE-only effect shows a larger response to climate change, increasing by 0.42 ppbv (10%) from +4.0 ppbv in 2000 climate to +4.4 ppbv in 2050 climate. The changes suggest that the 2000-2050 climate change will increase the sensitivity of surface ozone to a given change in domestic emissions by an annual average of 6% over China and 10% over CEC. This can be alternatively expressed as the sensitivity of the climate-only effects to domestic emissions. That is, Cases [(F-E) – (B-A)] = Cases [(F-B) – (E-A)]. The difference in the CHE-only effect between 2000 and 2050 climate is presented in Fig 6a for the summer condition. In the future climate, there is a significant increase (1-2 ppbv) in the

CHE-only effect on ozone over CEC and a decrease of up to 1 ppbv over West China. *Wu et al.* [2008a] suggested that the 2000-2050 change in global anthropogenic emissions would result in a decrease of the climate change penalty in the U.S. compared with the present day. This is because the A1B scenario assumes a 40% reduction in anthropogenic NO_x emissions in the U.S. in contrast to the 116% increase projected over China.

Wu et al. [2008a] proposed two mechanisms to explain why the sensitivity of ozone to domestic emissions depends on climate. First, the relative contribution of background ozone to surface ozone will change as a result of climate change. Second, the ozone production efficiency from anthropogenic NO_x emissions will depend on biogenic VOCs emissions, which in turn depend on climate [Lin et al., 1988; Sillman et al., 1990; Kang et al., 2003]. According to the two mechanisms, an increase of the same magnitude in CHE will result in a larger increase in surface ozone under the 2050 climate when biogenic VOCs emissions are higher and background ozone is lower, compared with the present-day climate. The policy implication is that a slower rate of increase in domestic emissions will be required to meet a given ozone air quality target in the 2050 climate over those regions where the sensitivity of ozone to domestic emissions will increase as a result of climate change.

As illustrated in Fig 6a, there is a decrease in ozone sensitivity to CHE over West China as a result of 2000-2050 climate change, which apparently cannot be explained by the above mechanisms. If we suppress the growth of biogenic emissions (all over the world) in the 2050 climate, the corresponding change in ozone sensitivity between the 2000 and 2050 climate is shown in Fig 6b. By comparing Fig 6a and 6b, we see that the increase of ozone sensitivity to CHE almost diminishes over CEC, while the decrease of sensitivity over West China still persists. This suggests that the coupling between increasing biogenic VOCs and anthropogenic NO_x emissions as suggested by Wu et al. [2008a] is the mechanism responsible for the increased ozone response to domestic emissions over CEC, but it is not the mechanism responsible for the decrease of ozone sensitivity to domestic emissions over West China. The mechanism for West China has to do with the coupling between climate and anthropogenic emissions which are the two factors included in the case of Fig 6b.

As the projected 2000-2050 changes of anthropogenic NO_x emissions are largest over east China, a question of interests is to what extent surface ozone over West China becomes less sensitive to anthropogenic emissions locally and those over CEC in the 2050 climate. We conducted another sensitivity simulation in which we suppress the growth of anthropogenic emissions over CEC region only. The corresponding change in ozone sensitivity between the 2000 and 2050 climate in this case is shown in Fig 6c. This case illustrates the change in the sensitivity of surface ozone to local emissions over West China in the future climate. We find that in this case, although the decrease of sensitivity still persists over West China (defined as the red rectangle in 6c), it becomes 40% smaller than that in Fig 6a. This suggests that compared with the present-day climate, surface ozone over West China in the 2050 climate is less sensitive not only to anthropogenic emissions locally but also to those over CEC, with the former contributing 60% of the overall change in sensitivity and the latter 40%.

Figure 7 displays the changes in surface NO_x mixing ratio as a result of the 2000-2050 changes in CHE (in the 2000 climate). The increase of surface NO_x mixing ratio is much smaller over West China than over CEC, making West China still in the low-NO_x region even with the 2050 Chinese emissions. As already shown in Section 3, the increase of water vapor in the 2050 climate will accelerate the destruction rate of ozone through reactions R2-R4 over low-NO_x regions, leading to decreased ozone lifetime and less efficient net ozone production from local emissions. In this situation, a given increase of local emissions over West China will result in a smaller increase of surface ozone in terms of absolute concentrations in the 2050 climate compared with the present-day climate. In addition, higher temperature in the 2050 climate will decrease the stability of PAN, resulting in less efficient export of NO_x from source regions in the east to West China. Both factors imply that there will be reduced sensitivity of surface ozone over West China to the overall emission changes in China in the future climate as shown in Fig 6a.

5.2. Sensitivity of RWE-only effect to climate

The RWE-only effect on surface ozone over China shows an annual decrease of 0.06 ppbv from the 2000 climate (Case C-A) to 2050 climate (Case G-E) [c.f. Fig 1].

Although the annual national-mean change appears small, it is important to understand the direction of the change and the spatial distribution. Figure 8 displays the the difference in the RWE-only effect between the 2000 and 2050 climate in the summer, the season of strongest ozone production. The impact of climate change on the RWE-only effect is negative all over China. This suggests that the 2000-2050 climate change will decrease the sensitivity of surface ozone to a given change in anthropogenic emissions from the rest of the world, although the magnitude of the change is small. The 2000-2050 climate change will result in a shorter lifetime of ozone and PAN due to higher temperature and more water vapor in the 2050 climate, as discussed before. The reduction in ozone lifetime will lead to less efficient export of ozone produced outside to China, causing a decrease in anthropogenic background ozone over China.

6. Combined effect of global change on surface ozone

Following the typical GCM-CTM approach, the differences between Case A (2000 climate and emissions) and Case H (2050 climate and emissions) in Fig 1 represent the response of surface ozone to the combined effect of 2000-2050 changes in climate and anthropogenic emissions of ozone precursors. Figure 9 shows the simulated changes in surface ozone over China in 2050 relative to the present-day case (Fig 2a). As a result of the 2000-2050 changes in both climate and anthropogenic emissions, annual mean surface ozone will increase by 8.7 ppb over China (Fig 9a), compared with the mean global change of 4.6 ppbv (not shown). Over CEC where the present-day ozone levels are already high, the increase in mean summertime surface ozone will reach an average of 15.9 ppbv (Fig 9b). The large increase of surface ozone over CEC will have important implications for public health and ecosystem.

As discussed before, the numbers shown along X, Y, and Z axes in Figure 1 are mean differences in surface ozone over China associated with the CHE-only, RWE-only, and climate-only effects respectively. To take into account the influence of one effect on the other, the average of all the CHE-only effects under different climate and RWE (i.e., along the four X-axes in Fig 1) is used to represent the contribution of the 2000-2050 CHE changes alone on overall ozone change. Similarly, the average of all the RWE-only effects along the four Y-axes represents the contribution of the

2000-2050 RWE changes and that along the four Z-axes the climate change contribution. Table 2 summarizes the mean CHE-only, RWE-only, climate-only, and the combined effects of all the factors over China and CEC. For China as a whole, the largest impact on surface ozone is from the projected change in RWE in the future, which contributes 64.6% of the total increase of annual-mean surface ozone in 2050. The projected change in CHE contributes 37.4% and climate change exerts a small negative influence of about -1.9%. For the heavily populated CEC region, the contribution from the change in Chinese emissions is the largest, responsible for 59.0% of the total effect. The contribution from RW emissions takes the second place of 43.2%. Most importantly, climate change represents a large positive effect (7.9%) over this region, resulting in a significant climate-change penalty. As we suggest above, an aggressive NO_x-emission control strategy is required to cope with this trend.

7. Concluding Remarks

We investigated the effects of 2000-2050 global changes on China's ozone air quality using a global chemical transport model (GEOS-Chem CTM) with meteorological inputs provided by a general circulation model (GISS GCM 3). The global anthropogenic emissions of ozone precursors are based on the IPCC A1B scenario. We decomposed future global changes into three factors: climate, Chinese anthropogenic emissions (CHE) and anthropogenic emissions from the rest of the world (RWE). In order to evaluate the effect of individual factors and the interaction between them, we constructed 8 different climate/CHE/RWE cases and for each case model simulations were performed for continuous three years to eliminate the interannual variation. The specific roles of biogenic emissions of VOCs and anthropogenic NO_x emissions were investigated through sensitivity simulations in some future scenarios.

In the 2050 climate, the annual mean temperature will increase 1- 2 K over most China, especially in the southeastern parts. A significant increase of humidity is found all over China, with the largest increases taking place in coastal areas. PBL heights decrease in central east China (CEC) but increase slightly in other regions. Under the present-day emissions, climate change will result in an average decrease of 0.21 ppbv in annual mean surface ozone for China, but an increase of 0.56 ppbv is found over CEC.

The increase over CEC is spatially extensive in summer, reaching up to almost 5 ppbv in some regions, posing a significant climate change penalty for public heath and ecosystem in this populated region with intensive agriculture. Further investigation shows that 43% of the climate change penalty over CEC could be attributed to the increase in biogenic emissions of VOCs. By contras, in western China climate change will decrease the ozone burden as a result of decreases in background ozone from elevated water vapor.

The changes in Chinese emissions alone will result in an increase of 3.2 ppbv in annual mean afternoon ozone over China and an increase of 4.0 ppbv over CEC. A significant increase of 9.0 ppbv is found over CEC in summer, suggesting the need for a more aggressive control strategy of Chinese emissions in order to mitigate ozone pollution in the future. More detailed investigation shows that the projected increase in anthropogenic NO_x emissions alone may explain 86% and 80% of the CHE-driven ozone increase over China and CEC respectively. This indicates that NO_x emission control should be the first priority in the emission control strategies.

The changes in RWE alone will result in an increase of 5.7 ppbv and 3.7 ppbv in annual-mean afternoon ozone for China and CEC respectively. The west-east and south-north gradient of surface ozone over China will decrease in the future, reflecting the relatively larger impact from ozone precursors emitted from neighboring countries to the south and west of China. In the A1B emission scenario, anthropogenic emissions of ozone precursors are projected to decrease in North America and Europe, offset by soaring increases of emissions from India and Southeast Asia. As a result, surface ozone levels over southwest China and the Tibet Plateau will be heavily affected and an increase of 10-15 ppbv in the summer is found for surface ozone over this region. It becomes crucial to establish an effective trans-boundary pollution control policy involving China, India and Southeast Asia countries in order to protect the fragile ecosystems and glacier over this region.

As a result of the combined effects of 2000-2050 changes in climate and emissions, annual mean surface ozone will increase by an average of 8.7 ppbv over China. Considering all the 8 climate/CHE/RWE cases, we found that the 2000-2050 change in RWE is responsible for 64.6% of the overall ozone change in China with domestic emissions contributing to 37.4% and climate changes -1.9%. The 2000-2050 global changes will result in an annual-mean ozone increase of 9.0 ppbv over CEC where Chinese emissions make the largest contribution (49.0%), followed by RW emissions (43.2%) and climate (7.9%). In contrast to the overall benefit of climate change on surface ozone over China, the climate change over CEC will result in a significant penalty on ozone, with an annual mean enhancement of 0.71 ppbv and summer-mean enhancement of 2.1 ppbv over this populated region. This indicates that climate change will aggravate ozone air quality over CEC in the future and pose further difficulty to mitigate ozone pollution given the increases in anthropogenic emissions in the future.

We find that 2000-2050 climate change will increase the sensitivity of surface ozone to domestic anthropogenic emissions by 10% over CEC, but decrease this sensitivity over west China. The former can be explained by the combined effects of increasing anthropogenic NO_x emissions and increasing biogenic emissions of VOCs, while the latter is attributed to the accelerated ozone destruction rate and reduced transport from CEC in the future climate. In contrast, the 2000-2050 climate change will decrease the sensitivity of surface ozone to RWE uniformly over China. This can be explained by a decrease in ozone lifetime in the future climate resulting in less efficient transport of anthropogenic background ozone to China. The increased sensitivity to CHE in the 2050 climate implies that domestic emission controls over China need to be more aggressive in the future.

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Tables

Table 1: The 2000-2050 trends in anthropogenic and natural emissions of ozone precursors.

		World			China		
		2000	2050	Change (%)	2000	2050	Change (%)
NOx（Tg N a^{-1})	aircraft	0.51	0.51	0.0	0.03	0.03	0.0
	Fossil fuel	24.21	43.25	78.6	3.35	7.24	116.4
	Biomass Burning	5.25	6.36	21.1	0.16	0.13	-17.5
	Biofuel	2.21	2.10	-4.6	0.65	0.46	-30.2
	Fertlizer	0.47	0.91	96.5	0.10	0.11	11.2
	lightning	4.92	5.79	17.6	0.35	0.44	25.7
	soil	6.73	7.69	14.3	0.52	0.58	11.9
CO (Tg a^{-1})	Biomass Burning	381.54	610.28	60.0	15.51	19.36	24.8
	Biofuel	175.15	168.33	-3.9	55.01	37.86	-31.2
	anthropogenic	330.95	397.74	20.2	100.57	90.27	-10.2
	monoterpenes	48.52	68.97	42.1	2.24	3.09	38.0
VOCs (Tg C a^{-1})	anthropogenic	48.7	127.8	160.2	7.86	15.92	102.4
	Biomass Burning	12.52	21.37	70.7	0.61	0.82	35.9
	Methane (ppb)	1750	2400	37.1			
Isoprene (Tg C a^{-1})		347.47	492.5	41.7	11.31	15.63	38.2
Other biogenic NMVOCs (Tg C a^{-1})		155.34	213	37.1	6.33	8.6	35.8

Table 2. The effect of the 2000-2050 global changes on annual afternoon-mean surface ozone over China and Central East China

	China		Central East China (CEC)	
Factors	ppbv	Percentage (%)	ppbv	Percentage (%)
2000-2050 change in China emissions[a]	3.3	37.4	4.4	49.0
2000-2050 change in world emissions (excluding China)[a] emissions	5.6	64.6	3.90	43.2
2000-2050 change in climate	-0.2	-1.9	0.7	7.9
Combined[c]	8.7	100	9.0	100

[a] Referred to as CHE in the text;
[b] Referred to as the rest of the world emissions (RWE) in the text;
[c] Here it means the combination of the 2000-2050 changes in the three factors (CHE, RWE, and climate)

Figures

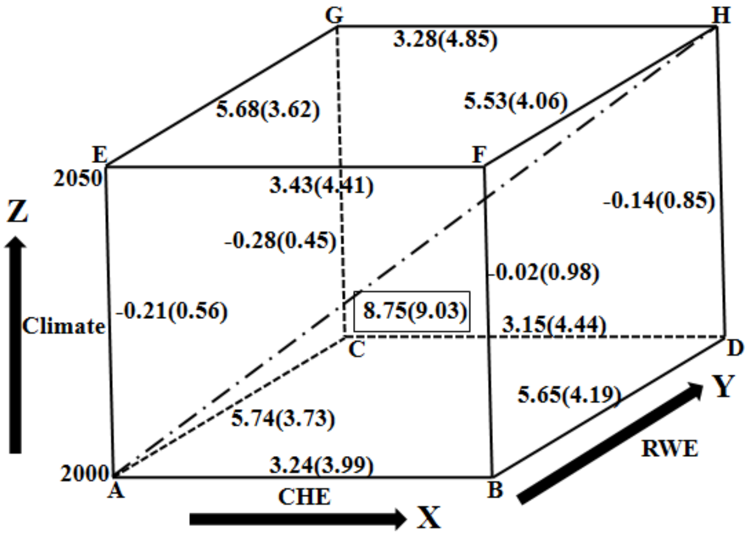

Figure 1. Schematic summary of the eight climate/emission sensitivity cases. The X-axis denotes the 2000-2050 changes in Chinese anthropogenic emissions (CHE), Y-axis the changes in anthropogenic emissions from the rest of the world (RWE), and Z-axis climate change. The numbers (unit: ppbv) displayed along the axes represent the differences between the cases in annual-mean afternoon ozone at the surface averaged over China and over Central East China (in parenthesis). The dash-dot line represents the direction of the change from 2000 climate and emissions to 2050 climate and emissions, and the boxed values are the associated ozone changes.

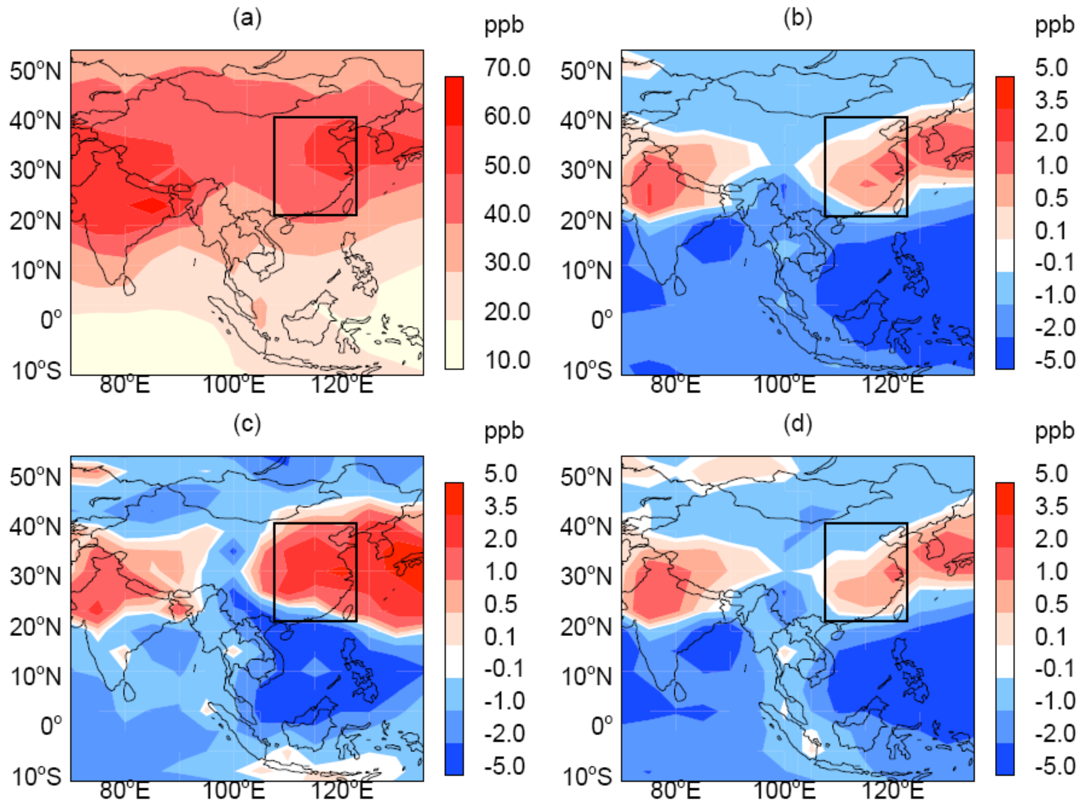

Figure 2. (a) The annual-mean distribution of surface ozone over China in the present-day climate and emissions (case A); (b) The climate-only effect on annual-mean surface ozone (case E minus case A); (c) same as b, but for the summer-mean; (d) same as b, but without the increase in biogenic emissions of VOCs in the 2050 climate. The black rectangle in each panel indicates Central East China (CEC).

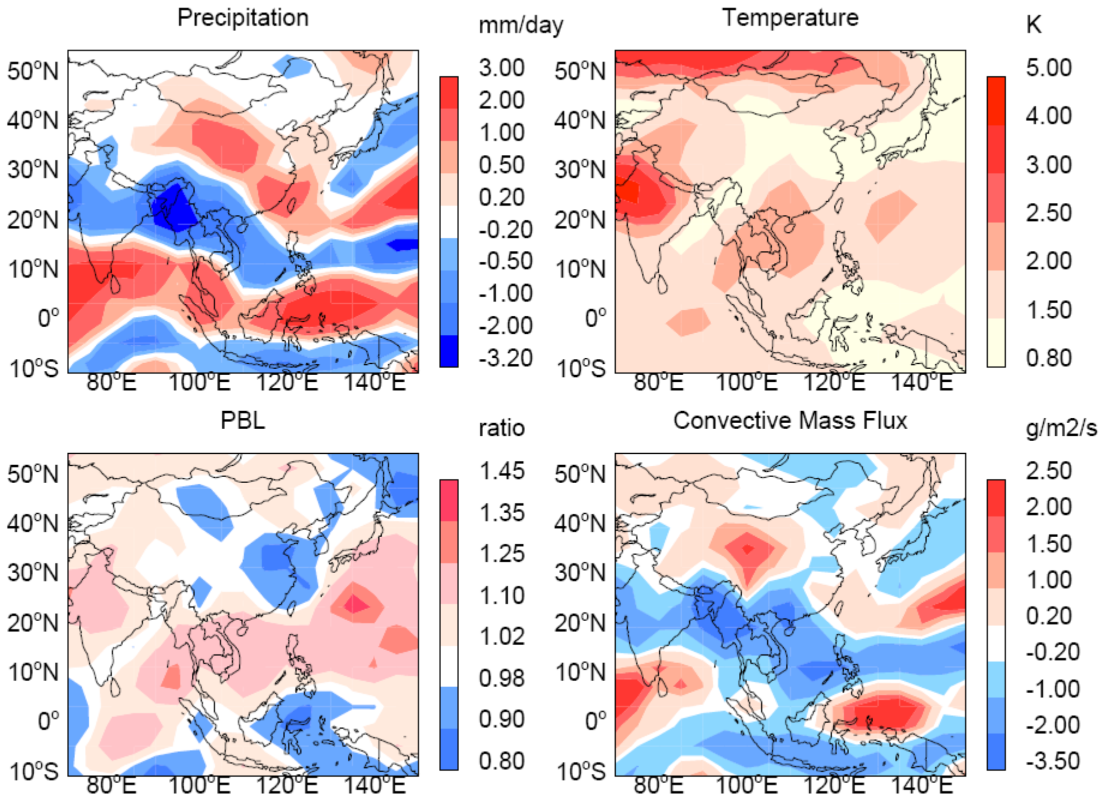

Figure 3. The 2000-2050 change in summertime (a) precipitation, (b) temperature, (c) planetary boundary layer depth (PBL), and (d) convective mass flux.

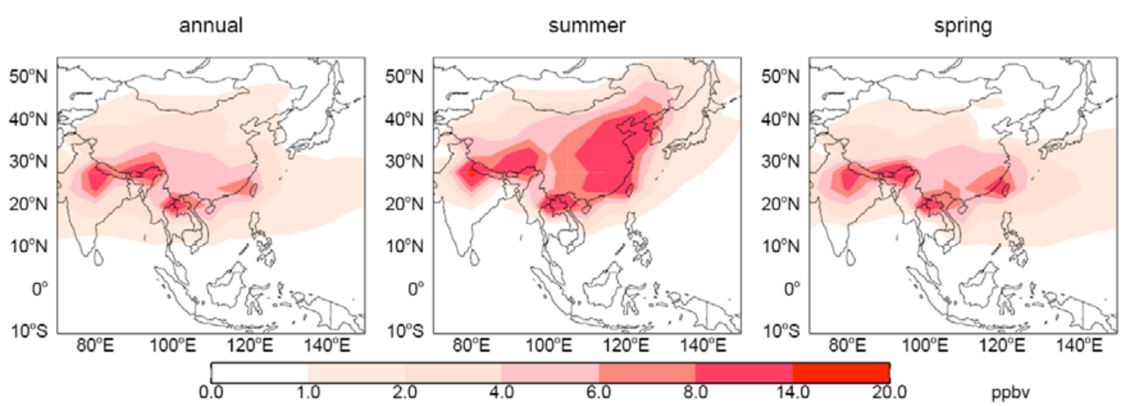

Figure 4. The effect of the 2000-2050 changes in Chinese anthropogenic emissions only on surface ozone: (a) annual-mean, (b) summer; (c) spring.

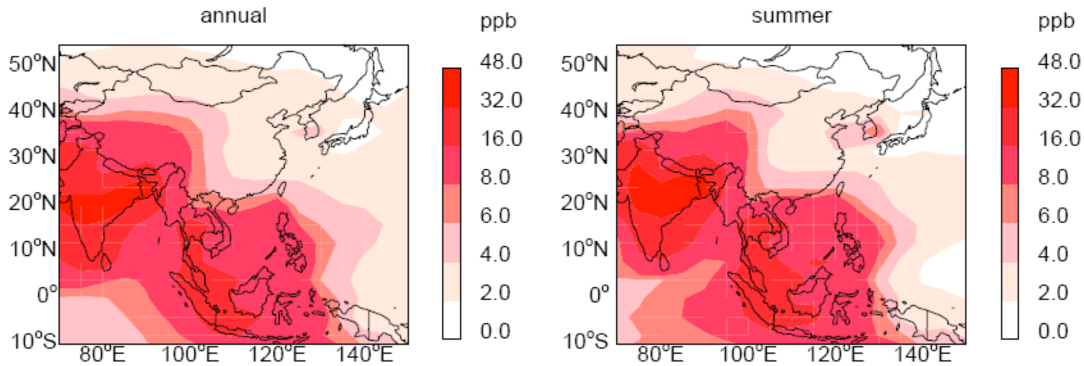

Figure 5. The effect of the 2000-2050 changes in anthropogenic emissions from the rest of the world on surface ozone: (a) annual mean; (b) summer.

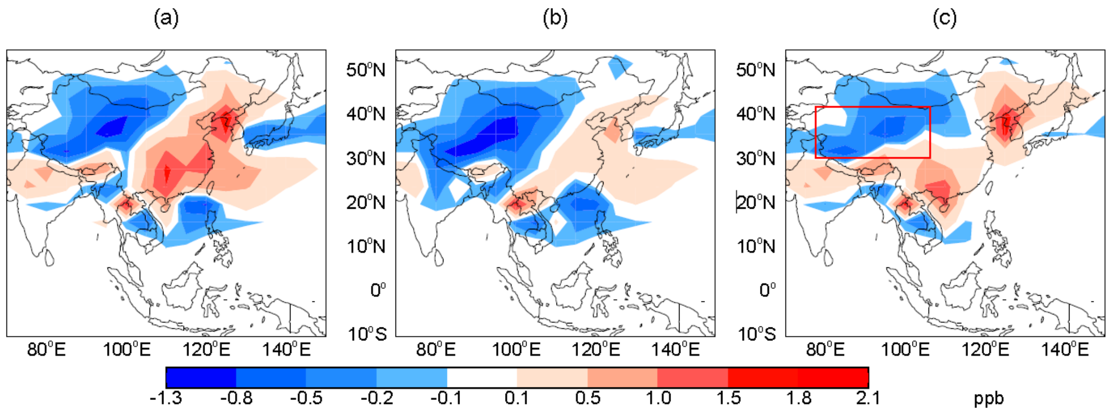

Figure 6. (a) The summer-mean difference in the effect of the 2000-2050 Chinese emission change on surface ozone between the 2000 and 2050 climate; (b) same as a, but without the changes in biogenic emissions of VOCs as a function of climate change; (c) same as a, but without the changes in the anthropogenic emissions over Central East China (CEC). The red rectangular region in (c) indicates West China.

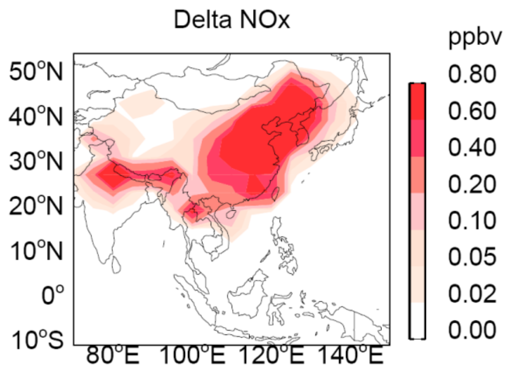

Figure 7. The effect on annual-mean surface NO_x mixing ratios in the 2000 climate brought merely by the 2000-2050 changes of Chinese anthropogenic emissions.

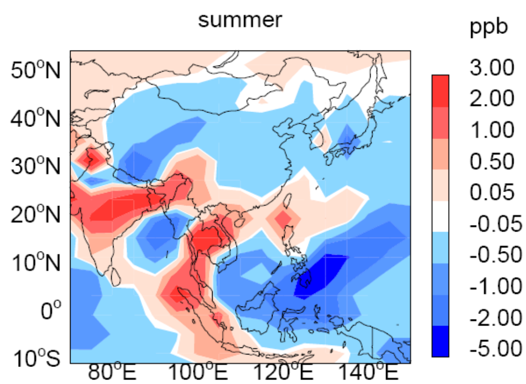

Figure 8. The summer-mean difference in the effect of the 2000-2050 global emission change (excluding China) on surface ozone between the 2000 and 2050 climate.

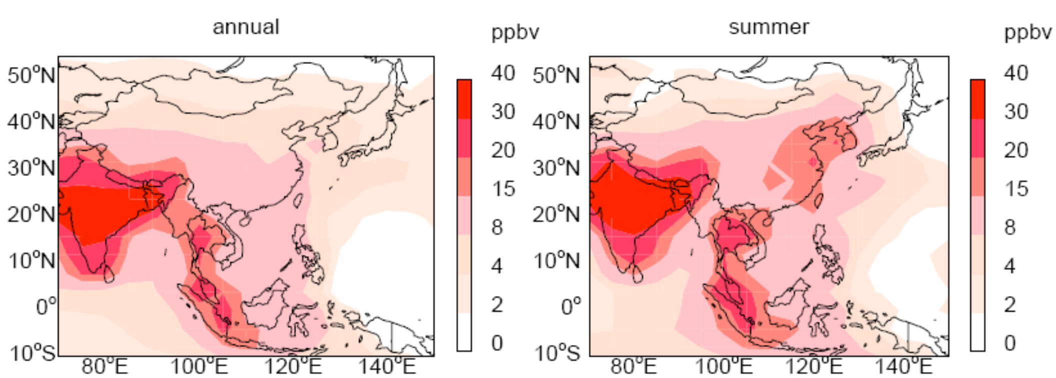

Figure 9. The overall change in (a) annual mean and (b) summertime surface ozone as a result of 2000-2050 global changes in both climate and emissions.