**Improving the Characterization of Pollution Transported into Texas**
**Tracking Number 2010-66, 2010-67**
**Grant Activities No. 582-8-86248-FY10-13**

**Final Report**

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1 June 2011

## 1. Abstract.

Using the chemical transport model GEOS-Chem, we have provided chemical boundary conditions for regional model simulations of U.S. air quality. We calculated the chemical composition of the global atmosphere for two scenarios: (1) 2005-2006 meteorology with 2005-2006 anthropogenic emissions and (2) 2005-2006 meteorology with 2018 emissions. We based the 2018 emissions on the A1B scenario from the Intergovernmental Panel on Climate Change (IPCC). This scenario describes a world of moderate socioeconomic growth and technology driven by a balance of fossil-fuel and alternative energy sources.

In 2018 in the A1B scenario, we found that global mean anthropogenic emissions of nitrogen oxides (NOx) increase by 9% relative to the year 2005, CO emissions by 20%, and emissions of volatile organic compounds (VOCs) by ~16-50%, depending on species. The global burden of methane increases by 12%. Emission changes are largest in Asia, especially in India. Over the United States, A1B anthropogenic emissions decrease by 5% for NOx, 14% for CO, and 5-10% for most VOCs, relative to the year 2005.

Results show that the 2018 change in U.S. emissions leads to a ~1 ppb decline in summertime (JJA) surface ozone concentrations in the East, relative to 2005-2006. JJA surface ozone concentrations in the western half of the United States, however, increase by 1-2 ppb in the 2005-2018 timeframe due to the increased ozone burden in the background atmosphere. The ozone increase in the West occurs despite the widespread 10% decrease in surface NOx concentrations across the United States. Our results point to the importance of considering foreign emission sources when planning for improvement of U.S. air quality.

## 2. Introduction.

The development of State Implement Plans to improve U.S. air quality requires knowledge of trends in precursor emissions outside the United States as well as within the nation. Also needed is confidence in the regional models used to predict surface levels of pollution. This project was undertaken in support of the efforts of the Texas Commission on Environmental Quality (TCEQ) to improve the characterization of long-range transport of ozone and ozone precursors in regional air quality models. The project involved use of GEOS-Chem, a global atmospheric chemistry model, to provide boundary conditions for the TCEQ regional models. GEOS-Chem was used to simulate the present-day (2005-2006) atmosphere, as well as projections of a future (2018)

atmosphere, characterized by changing emissions of anthropogenic ozone sources.

Tropospheric ozone is produced via the photochemical oxidation of volatile organic compounds (VOC) and carbon monoxide (CO) in the presence of nitrogen oxides (NOx). Model studies suggest that background levels of ozone can contribute considerably to US regional air quality [Fiore et al., 2002a; Fiore et al., 2000b; Hudman et al., 2004]. For example, Fiore et al. [2002a] found that present-day anthropogenic emissions of ozone precursors in Asia and Europe enhance afternoon ozone concentrations in surface air over the United States by 4–7 ppb. Observations from the western United States appear to confirm these model studies [Jaffe et al., 2003; Cooper et al., 2010].

In the coming years, emissions of ozone precursors are expected to rise in developing countries, especially in Asia [Streets et al., 2004; IPCC, 2007], with probable consequences on U.S. air quality. To gauge the importance of remote pollution sources on ozone air quality in the United States, many model studies have performed sensitivity studies, applying incremental increases (or decreases) to the emissions of ozone precursors abroad. In a multi-model study, Fiore et al. [2009] calculated that a 20% decrease in the foreign emissions of NOx, VOCs, and CO would reduce North American ozone levels by about 0.3 ppb in summer. In another sensitivity study, Fiore et al. [2002b] calculated that a 50% decrease in global anthropogenic methane emissions would have nearly as great an impact on mean afternoon surface ozone concentrations across the United States as a 50% decrease in global emissions of NOx. Fiore et al. [2009] revisited this result and found that a change in anthropogenic methane emissions from a foreign source region yields an ozone response in a receptor region that roughly equals that produced by combined change of the same relative magnitude in anthropogenic NOx, VOCs, and CO emissions [Fiore et al., 2009].

Recent model studies have also attempted to capture the response of U.S. ozone air quality to realistically changing foreign sources. For the time period 1996 to 2002, current models estimate an increasing trend of +0.1 ppb $a^{-1}$ in background ozone over the United States, at the low end of the 0.1–0.5 ppb $a^{-1}$ derived from observations [Fiore et al., 2009]. For the future near-term (present-day to 2030), the impact of future trends on U.S. surface ozone varies dramatically according to scenario. In the worst case A2 IPCC scenario, in which developing countries rely heavily on fossil fuel sources of energy, annual mean ozone concentrations increase by 4-6 ppb across the United States by 2030 [Dentener et al., 2006]. Even in the more moderate A1B scenario, Fiore et al. [2002b] calculated a longer and more intense U.S. ozone pollution season in 2030 relative to 1995, despite domestic emission reductions. On the other hand, in the Maximum Feasible Reduction scenario, U.S. ozone levels are projected to drop by an 4-6 ppb [Dentener et al., 2006].

Results from the present project will add to this body of knowledge regarding the impact of emissions abroad on U.S. air quality.

## 3. Project accomplishments.

### 3.1 Present-day anthropogenic emissions.

The present-day anthropogenic emissions in GEOS-Chem are based on the global inventory from GEIA [Benkovitz et al., 1996]. Various adjustments are made to this base. To provide a more current estimate of global emissions of NOx, SOx and CO, including values from ship emissions, we implemented the EDGAR 3.2FT2000 emissions [Olivier et al., 2001]. Over the United States, we applied the EPA NEI emissions; over Mexico, the BRAVO emissions [Kuhns et al., 2005]; and over Canada, the CAC emissions. For eastern Asia, we used the inventories of Streets et al. [2003, 2006] and Zhang et al. [2006]. For Europe, we applied EMEP emissions [Vestreng et al., 2007]. Black carbon and organic carbon aerosol are from Bond et al. [2004], overwritten by Cooke et al. [1999] for the United States.

To generate the 2005 emissions for the TCEQ project, we scaled all regional and global inventories from their respective base year to 2005. Our approach follows Bey et al. [2001] and Park et al. [2004]. Emissions are scaled according to estimates provided by individual countries or regions, where available. These countries and regions include the United States, Canada, Japan and Europe. Over Southeast Asia, we used the scale factors from Ohara et al. [2007]. The anthropogenic emissions from other countries are scaled according to the local trends in $CO_2$ emissions. NOx emissions are scaled by the relative change in total $CO_2$ emissions; SOx emissions by the change in solid fuel $CO_2$ emissions; and CO emissions by liquid fuel $CO_2$ emissions. The $CO_2$ emission data are from the Carbon Dioxide Information Analysis Center. Finally, we also applied diurnal scale factors to NOx emissions, using information from the EDGAR inventory.

### 3.2 Future anthropogenic emissions.

To calculate the 2018 emissions, we relied in large part on the IPCC decadal projections for emissions of greenhouse gases, aerosols, and ozone precursors [Nakicenovic et al., 2000]. These projections describe four socioeconomic scenarios (A1, A2, B1, B2). The A1 scenario further distinguishes three subscenarios (A1FI, A1T, A1B) by technological emphasis. All the IPCC scenarios predict global increases of anthropogenic emissions of ozone precursors for 2000–2050, largely driven by economic growth in developing countries. We chose to apply the A1B scenario for the 2018 emissions. This scenario describes a world of moderate socioeconomic growth. Sources of energy in the A1B scenario consist of a balanced mix of fossil-fuel and alternative technologies.

For speciation of the future VOC emissions and for updated projections of NOx emissions, we relied on Streets et al. [2004]. Streets et al. [2004] also allocated the trends in all anthropogenic emissions over 17 regions and 8 economic sectors. In a previous project, we followed the approach of Streets et al. [2004] to generate scaling factors in all regions for a suite of emissions, including ozone precursors, for the year 2030 relative to 2000. To calculate emissions in 2018 for the TCEQ project, we assumed a linear growth in anthropogenic emissions from 2005 to 2030, and interpolated the Streets et al. [2004]

scaling factors to 2018. We then applied the 2018 scaling factors to 2005 emissions. Methane is a special case in the future scenario. Since methane is a long lived gas, we simply specified its concentration in GEOS-Chem, following the IPCC [2007] recommendation for the A1B scenario.

Using this approach, we found that global mean emissions of all anthropogenic precursors of ozone increased in the 2018 inventory, relative to that for 2005. As shown in Table 1, we calculated that global mean emissions of nitrogen oxides (NOx) increase by 9%, CO emissions by 20%, and emissions of volatile organic compounds (VOCs) increase by 16-50%, depending on species. The global burden of methane increases by 12%, from 1.76 ppm to 2.00 ppm. These trends, however, show large regional variations, as shown in Figure 1. Emissions in the developing world increase, while those of the developed world decrease. For example, NOx emissions in India and China increase by 20-50%. Africa sees large relative increases in emissions in the 2018 projections, but the absolute change is very small since the present-day emissions there are small. Mexico also experiences a large increase in NOx emissions of ~20%. Over the United States, however, A1B anthropogenic emissions decrease by 5% for NOx, 14% for CO, and 5-10% for most VOCs, relative to the year 2005 (Table 1).

Figure 2 shows the distribution of 2018 emissions for two important ozone precursors, CO and NOx, and provides a representative idea of emission hotspots in the A1B scenario. These hotspots include China, India, South Africa, and eastern Europe.

**Table 1.** Percentage changes in anthropogenic emissions from 2005 to 2018 conditions.

| Species | Global | United States |
|---|---|---|
| NOx | +9 | -5 |
| CO | +20 | -14 |
| $CH_2O$, aldehydes | +26 | -6 |
| $C_2H_6$ | +56 | -10 |
| C4, C5 alkanes | +29 | -9 |
| Propene | +31 | -15 |
| Propane | +51 | -5 |
| Acetone | +16 | +1 |

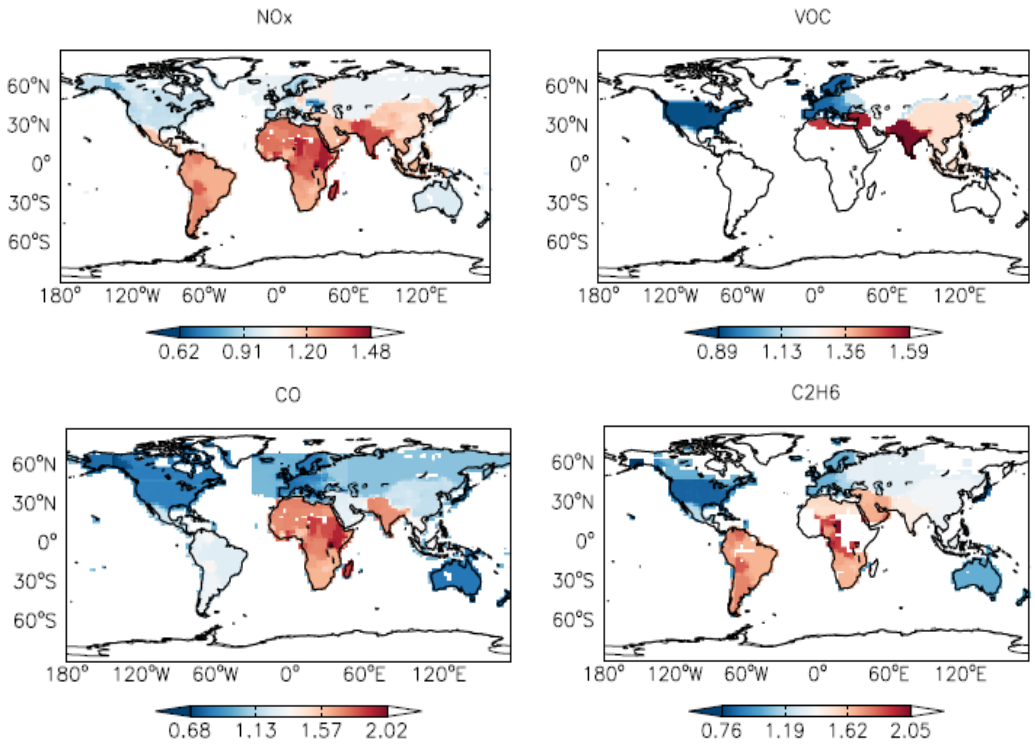


**Figure 1.** Fractional change in anthropogenic emissions for selected ozone precursors in the 2018 atmosphere, relative to 2005. Here VOC signifies formaldehyde and other aldehydes. Note differences in scales for the panels.

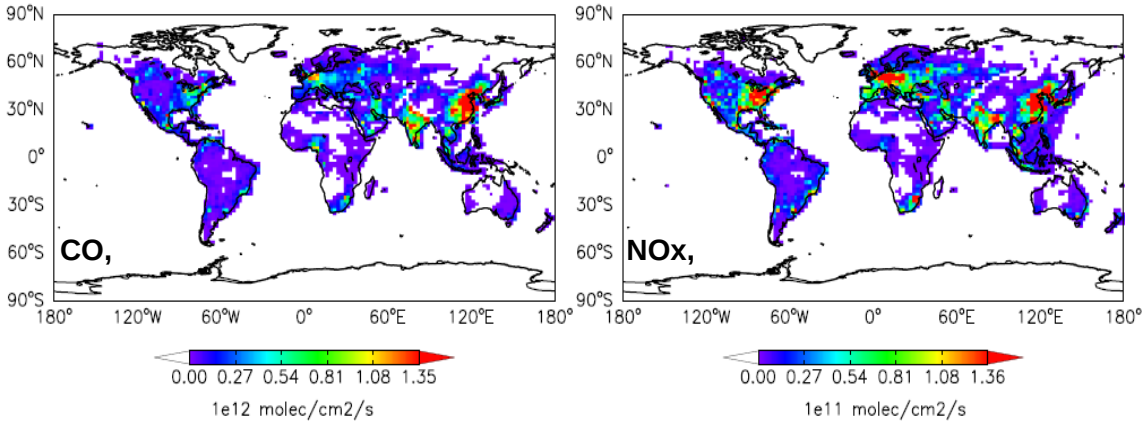


**Figure 2.** Calculated emissions for anthropogenic CO and NOx in the 2018 A1B scenario.

**3.3. Simulation of chemistry.**

For the chemistry simulations, we used the Harvard chemical transport model GEOS-Chem, version 8.03.02 with $2^o$ latitude x $2.5^o$ longitude horizontal resolution. Meteorological fields were from GEOS-5. We performed 4 years of simulations:

a. 2005 meteorology with 2005 base-year emissions
b. 2005 meteorology with 2018 future-year emissions
c. 2006 meteorology with 2006 base-year emissions
d. 2006 meteorology with 2018 future-year emissions

Present-day meteorology was used in all simulations in order to isolate the response of U.S. air quality to changing emissions. Application of present-day and future anthropogenic emissions was straightforward. To examine the impacts of changing emissions, we compared the mean concentrations of ozone and other pollutants calculated with 2005 and 2006 emissions (simulations a+c) with the mean concentrations of the two years calculated with 2018 emissions (simulations b+d).

For all four years, we archived timeseries of 54 tracers and grid box heights with one-hour resolution for the bottom 40 layers over the rectangular domain bounded by $136^o$ W and $59^o$ W and by $18^o$ N and $62^o$ N. These timeseries were saved as one file per model day, and will be shipped to TCEQ on a harddrive for use as boundary conditions in the TCEQ regional models.

**3.4. Impact of emission trends on surface ozone.**

**3.4.1. Outside the United States.** Not surprisingly, Asia is the region most affected by the 2005-2018 trends in emissions. Mean ozone concentrations over the Middle East, India, and China increase 3-9 ppb in summer (Figure 3). Over Mexico, surface ozone levels rise by 1-2 ppb from spring through fall, consistent with the projected increases in NOx emissions and other ozone precursors in that country.

**3.4.2. Within the United States.** Figure 4 shows the projected 2018 changes in surface concentrations for three important ozone precursors – NOx, $CH_2O$, and CO – in summer. NOx concentrations decrease ~10% across most of the United States. $CH_2O$ shows decreases of 5% in the Southeast, but increases of 5-10% elsewhere, while CO concentrations decrease strongly in the East and in polluted regions in the West. The increases in emissions in a few gridboxes in the northern Rockies is due to a small enhancement in 2018 biomass burning in this region. The consequences of increased emissions in Mexico is clearly visible in Figure 4, leading to a 20% enhancement in surface NOx concentrations and 10% enhancements in $CH_2O$ and CO.

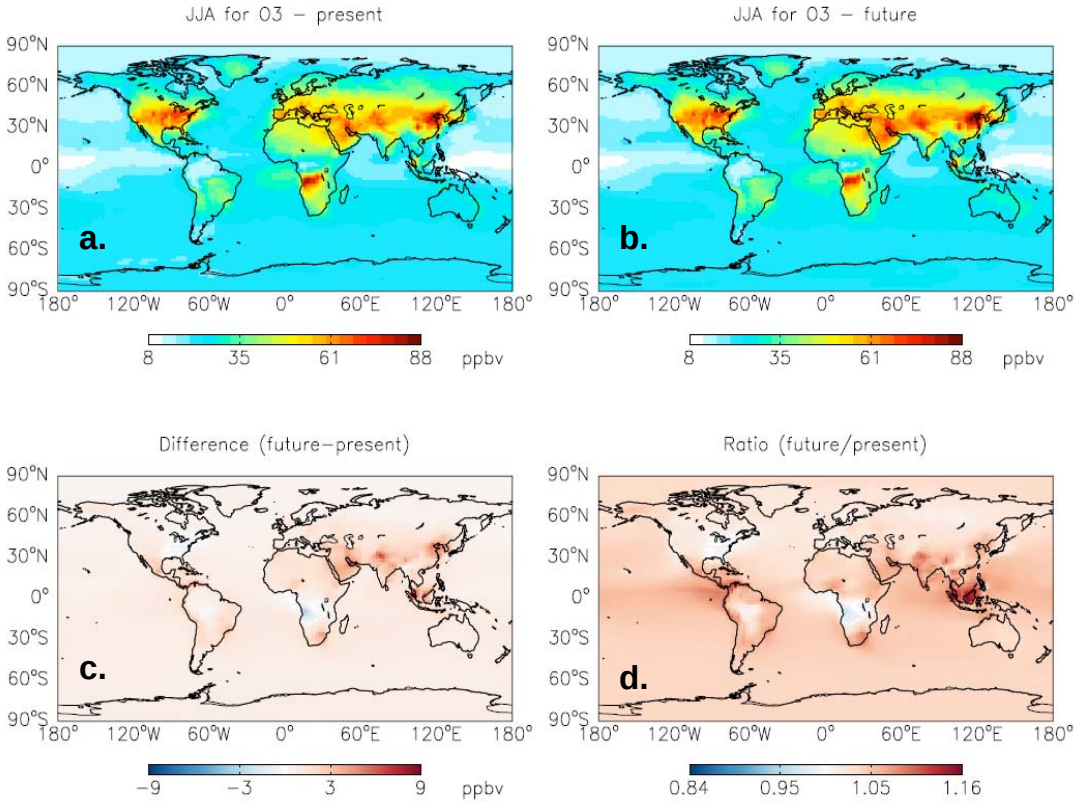


**Figure 3.** Global mean JJA afternoon surface ozone concentrations and the response to the 2005-2018 emissions trend: (a) present-day (2005-2006) surface ozone concentrations, (b) future surface ozone concentrations, calculated with 2005-2006 meteorology but with 2018 emissions, (c) difference between future and present-day ozone concentrations, (d) ratio of future to present-day ozone concentrations.

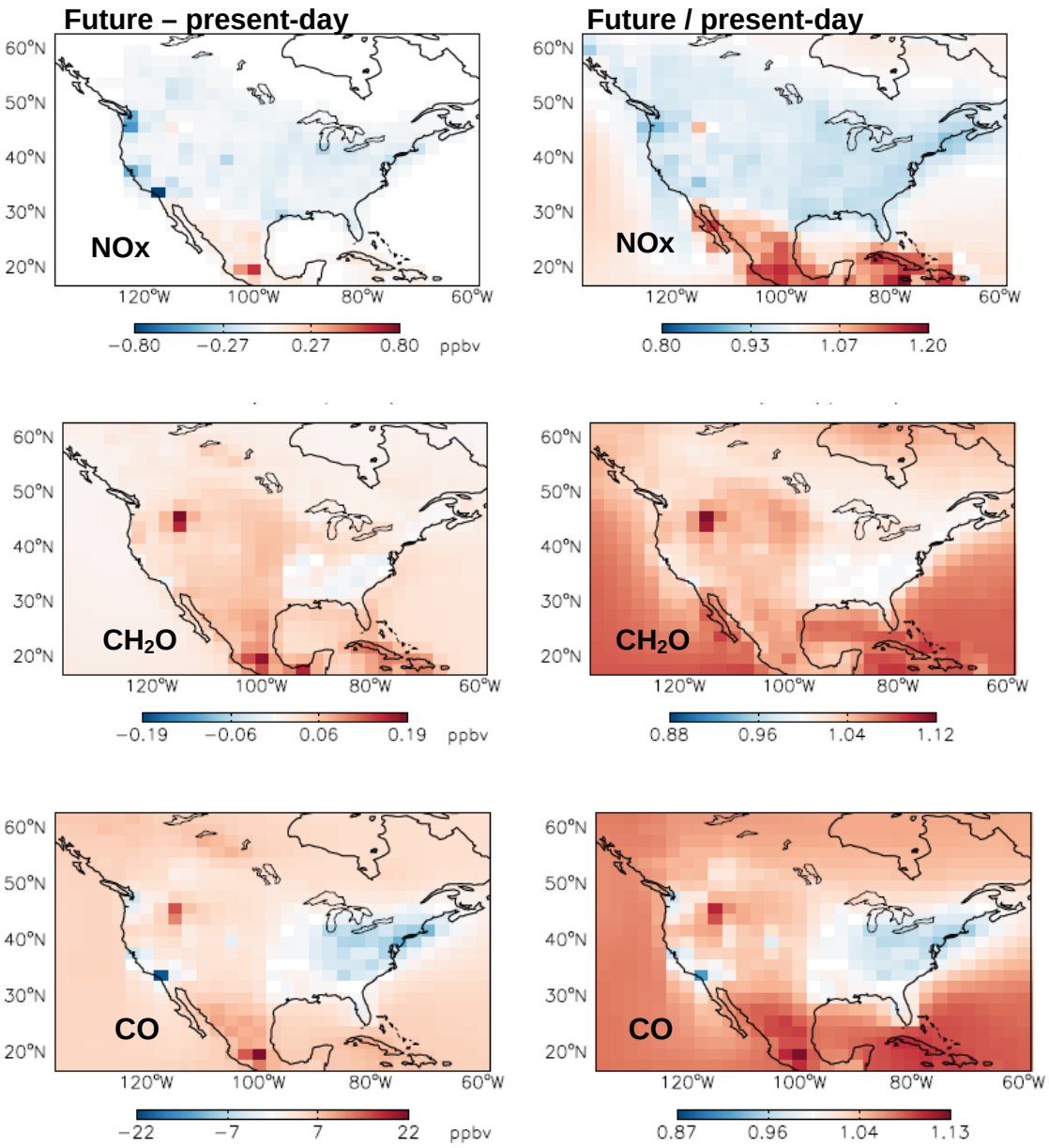


**Figure 4.** Differences between future and present-day surface concentrations of NOx, $CH_2O$, and CO in summer over the United States (left column). Future concentrations are calculated for the 2018 A1B atmosphere; present-day is taken as the 2005-2006 mean concentration. The right column shows the ratio of future / present-day concentrations.

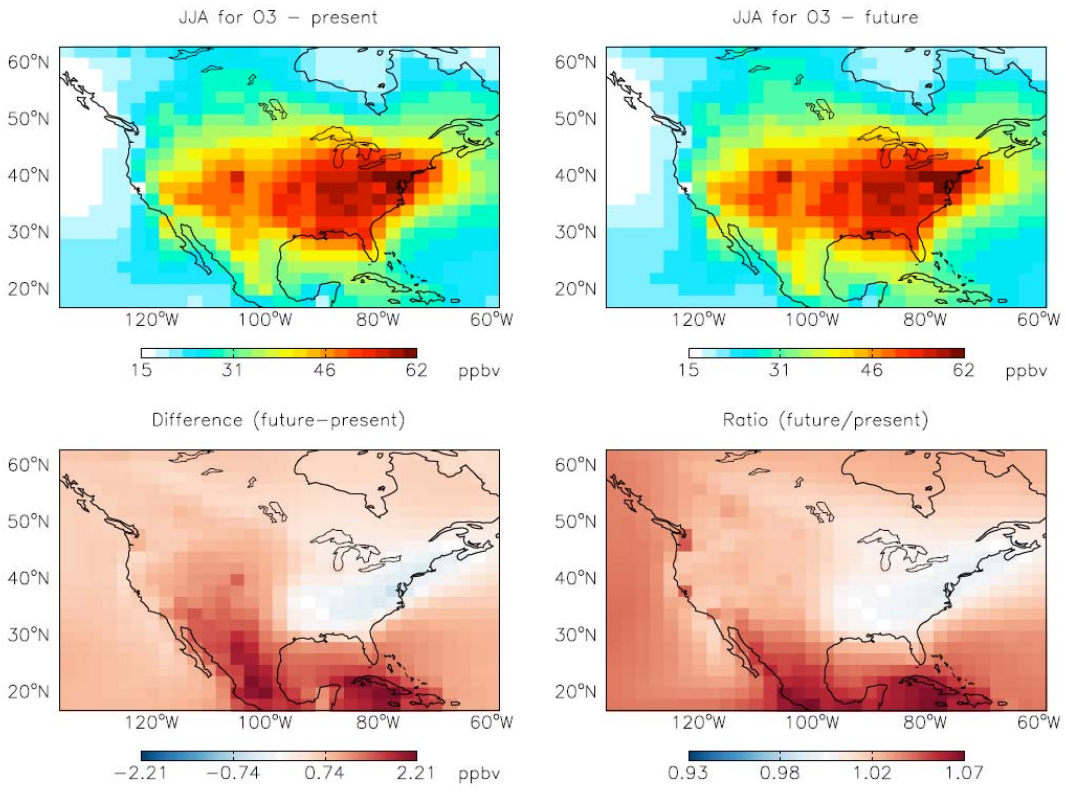


**Figure 5.** Mean JJA afternoon surface ozone concentrations and the response to the 2005-2018 emissions trend over much of North America: (a) present-day (2005-2006) surface ozone concentrations, (b) future surface ozone concentrations, calculated with 2005-2006 meteorology but with 2018 emissions, (c) difference between future and present-day ozone concentrations, (d) ratio between future and present-day ozone concentrations.

Figure 5 shows in more detail the 2005-2018 change in mean JJA afternoon surface ozone concentrations over North America. The eastern half of the United States experiences a decrease of ~1 ppb surface ozone due to the decrease in emissions of ozone precursors (Figure 4). The western half of the country, on the other hand, shows enhanced ozone of 1-2 ppb. These enhancements are due to transport of Asian pollution together with an increase in global ozone production from methane. The western United States is vulnerable to changes in background ozone due to its proximity to Asia and to strong subsidence in the Southwest [Fiore et al., 2002a]. By contrast, Mexico shows large increases in surface ozone, more than 2 ppb, because of the increase in local emissions.

These results, which show decreases in 2018 surface ozone concentrations in the eastern United States but increases in the West, are consistent with previous studies. For example, a recent multi-model study estimated that a 20% decrease in foreign ozone precursor emissions reduces North American ozone levels by about 0.3 ppb in summer [Fiore et al., 2009]. In our case, Asian emissions increase 10-50%, depending on species.

For the time period 1996 to 2002, current models have estimated an increasing trend of +0.1 ppb $a^{-1}$ in background ozone over the United States, or about 0.7 ppb total increase [Fiore et al., 2009]. Observations report an even larger increase of background ozone about 0.1–0.5 ppb $a^{-1}$ during this period, at least in the West [e.g., Jaffe et al., 2003; Parrish et al., 2004]. Looking ahead to 2050 in the A1B scenario, Wu et al. [2008] have calculated that the increase in global anthropogenic emissions of ozone precursors could increase U.S. background ozone by 2–6 ppb in summer, with the largest increases in the West. The extent to which these projected changes in background ozone will affect ozone episodes in the United States is an issue that will be addressed by the next phase of Texas project.

**3.5. Shortfalls and limitations of the project.**

A limitation of the project was that we did not perform sensitivity studies that would have allowed us to diagnose all the changes in surface ozone over the United States in the 2018 atmosphere. Some questions left unresolved are as follows:

- What is the relative importance of Asian vs. Mexican emissions regarding U.S. ozone air quality? Previous studies suggest that Mexican air quality has little influence on United States [Wang et al., 2009].

- How much does the increased 2018 global methane burden influence the background ozone levels in the United States, relative to foreign changes in emissions of NOx, VOCs, and CO?

- To what extent will projected changes in background ozone affect the frequency of ozone exceedances in the United States?

**4. Recommendations.**

We recommend that TCEQ, in applying our chemical boundary conditions to their regional models, work closely with Harvard to ensure that the chemical species are properly "translated" into the TCEQ chemistry scheme.

Also, in performing simulations of background ozone on Texas air quality, it would be of interest to know under what conditions the effect of background ozone is greatest. Previously, Fiore et al. (2002a) found that the background contribution is generally highest when surface ozone is in the 50–70 ppb range, and decreases when ozone is higher because of restricted import in stagnant air masses.

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