Supporting Information for

How have Divergent Global Emission Trends Influenced Long-range Transported Ozone to North America?

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**S1. Model Performance Evaluation**

**S1.1 Comparison with surface network measurements**

Extensive evaluation of surface-level predictions of O3 and NO2 (along with several other chemical species) from the Hemispheric CMAQ (HCMAQ) model simulations discussed in Section 2.1 for the 1990-2010 period have previously been conducted through comparisons with available measurements from surface networks in the United States, Europe, and Asia and are described in Xing et al. (2015). The evaluations utilized observations from two networks in the United States, i.e., Air Quality System, (US-AQS, <http://www.epa.gov/ttn/airs/airsaqs/>), and US-CASTNET; two networks in Europe, i.e., EU-EMEP and EU-AIRBASE; one in China, CN-API (Air Pollution Index), and one global network, WDCGG. The US-CASTNET and EU-EMEP networks are designed for air pollution trend assessment since their monitors are typically located in rural areas and represent regional air pollution. In contrast, sites in the US-AQS and EU-AIRBASE are closer to urban areas, impacted by local emissions and are typically representative of much smaller regions that are sub-grid to the 108km grid spacing utilized in the current simulations. The CN-API data are average values of observations from all sites within a city as detailed in Xing et al. (2015). Model trends were also assessed at three WDCCG sites in East Asia.

For each network, five statistical performance measures were used to evaluate the model predictions and included: correlation coefficient (R), mean bias (MB), normalized mean bias (NMB), root mean squared error (RMSE) and normalized mean error (NME) (see Table 2a in Xing et al. (2015)). Briefly, relative to the urban networks, NO2 was underestimated: US-AQS by 48 %, EU-AIRBASE by 54% and CN-API by 68%. This underestimation can be attributed to artificial dilution of NOx emissions over the relatively coarse model grid size and comparison with near source monitors. In contrast the performance for NO2 relative to measurements from rural networks was significantly better with biases within ±15% for all seasons. The NMB (-13.6 to 16.9 %) and NME (< 25.9 %) values for the daily maximum 8-hr average O3 predictions with measurements from the various networks in the U.S., Europe and Asia were fairly small across all seasons and comparable with those reported in other modeling studies in these regions (e.g., Zhang et al., 2009; Wang et al., 2009; Colette et al., 2011).

Simulated trends in surface-level ambient NO2 for the 1990-2010 period across the Northern Hemisphere exhibited spatial heterogeneity which followed the trends in regional NOx emissions. As reported in Xing et al. (2015), the annual change rates in simulated surface-level NO2 for China (4.1%), U.S. (-1.4%) and Europe (-1.2%) matched closely with the trends in input NOx emissions in these regions which were 4.3%, -1.8%, and -1.5%, respectively. The NO2 trends (and annual change rates) inferred from measurements available at the US-AQS and EU-AIRBASE networks were -0.63 µg m-3 yr-1 (-2.3%) and -0.64 µg m-3 yr-1 (-1.9%), respectively. The model estimated NO2 trends at these networks, -0.32 µg m-3 yr-1 (-2.2%) and -0.14 µg m-3 yr-1 (-0.9%), respectively, were however underestimated and are related to the underestimation of the NO2 concentrations. Additionally, comparison of 2003–2010 trends in tropospheric NO2 between the HCMAQ simulations and SCIAMACHY reported in Mathur et al. (2017) indicate that the model captures increases in eastern China, and many cities in India and the Middle East as well as the decreases across the eastern United States, southern California, and western Europe.

The decreasing trends in observed DM8O3 for US-CASTNET both on an annual and seasonal basis was captured by the model with strongest decreasing trend during summer (see Table 4 in Xing et al., 2015). However, the magnitude of the trend both on a networkwide basis (Xing et al., 2015) and at individual sites was underestimated by the model; the annual observed trend (and annual change rate) for DM8O3 were -1.859 µg m-3 yr-1 (-1.1%) while the model estimates were -0.952 µg m-3 yr-1 (-0.64%). Similar underestimation of the trends is also evident in the modeled 5th and 25th percentile O3 distributions at the surface illustrated in Figures 2a and 2d. However, as illustrated in Figure 15a of Mathur et al. (2017) this underestimation is significantly rectified in nested finer resolution simulations over the contiguous U.S. that utilized a 36 km horizontal resolution and an updated emission inventory for the U.S. for the 1990-2010 period. Since the nested simulations were driven by space and time varying lateral boundary conditions from the HCMAQ (Gan et al., 2015) and since the forcing in the upper layers of the regional model is largely driven by the large scale forcing derived from the hemispheric simulation (as demonstrated in Hogrefe et al., 2018), both the nested model and the HCMAQ model can be expected to show trends aloft similar to those in Figures 2b,c,e,f. The improved magnitude of the trend and the improved correlation in the nested finer resolution model at surface monitors is suggestive of local influences better resolved by finer resolution and improved emission estimates since the long-range transport impacts are the same in both models. Since the emphasis of the current study is this large scale forcing and long-range transport aloft, and since these show an increasing trend aloft, the impact of which on surface-level O3 is better captured in the nested finer resolution calculations, the noted underestimation of the magnitude of the trend in the hemispheric coarser grid model calculations relative to surface monitors should not adversely impact conclusions related to the relative importance of the different source regions to long-range transported O3.

**S1.2 Comparison with surface measurements at Trinidad Head**

O3 measurements at Trinidad Head, California (41° 3´N, 124° 9´W) have often been used to represent the amount of O3 in air masses advected over the Pacific and entering the Western U.S. Several studies have analyzed the O3 measurements at the Trinidad Head surface site to characterize variations and changes in “baseline” ozone, defined as measurements in air without recent anthropogenic and continental influences (e.g., Parrish et al., 2020). Figure S1 presents comparisons of simulated seasonal mean surface-level O3 values for the HCMAQ grid-cell in which the Trinidad Head site is located, with corresponding measurements for the years 2002-2010 which is the common time period for available measurements and the multi-decadal simulations considered in the analysis in the main text. To isolate baseline conditions, measurement-based analyses have often focused on periods of strong onshore winds to eliminate local and continental influences. However, since our model grid resolution is relatively coarse and since the grid cell in which the Trinidad Head site is located has a 37% water and 63% land coverage, we cannot eliminate influences of local emissions from portions of the grid cell with land area or uniquely screen for baseline conditions as in prior measurement studies.

Chart, scatter chart

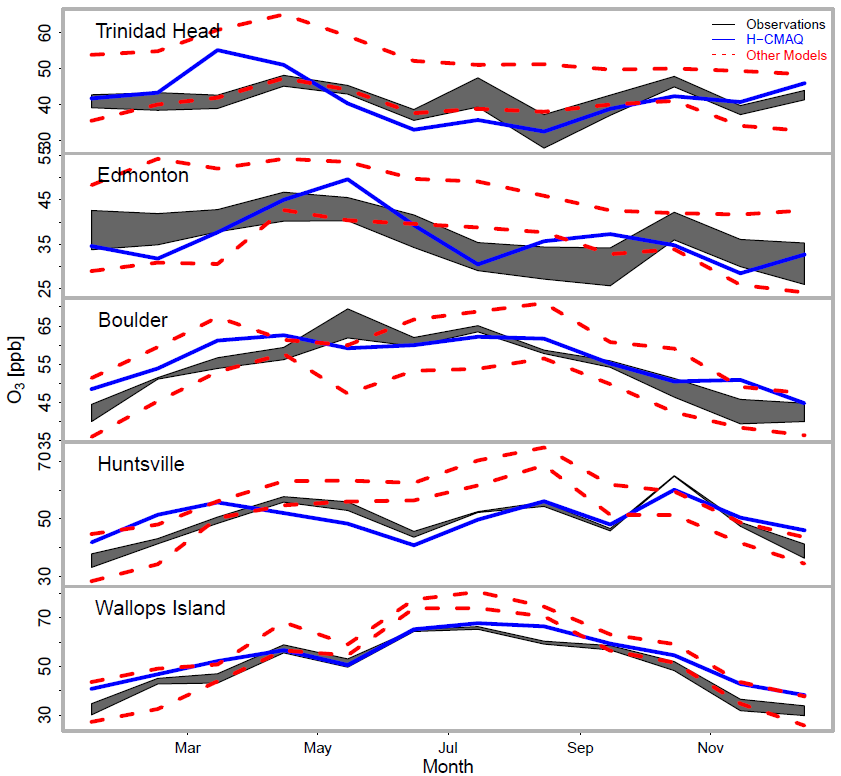
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**Figure S1**: Comparison of model and observed seasonal mean ozone mixing ratios at Trinidad Head for the years 2002-2010. Dash lines indicate that the model values are within 25% of the observed.

As illustrated in Figure S1, the model exhibits a seasonal cycle with high values in Spring, low in summer which then increase in Fall, consistent with the observations though the model does exhibit a systematic high bias during the Fall. Despite the limitations of the coarse model resolution (and inherent land/water coverage) in representing marine boundary layer conditions, the model predicted seasonal mean values at the lowest model layer are well within 25% of the observed surface measurements at Trinidad Head.

**S1.3 Comparison with other global models and aloft** O3 **measurements**

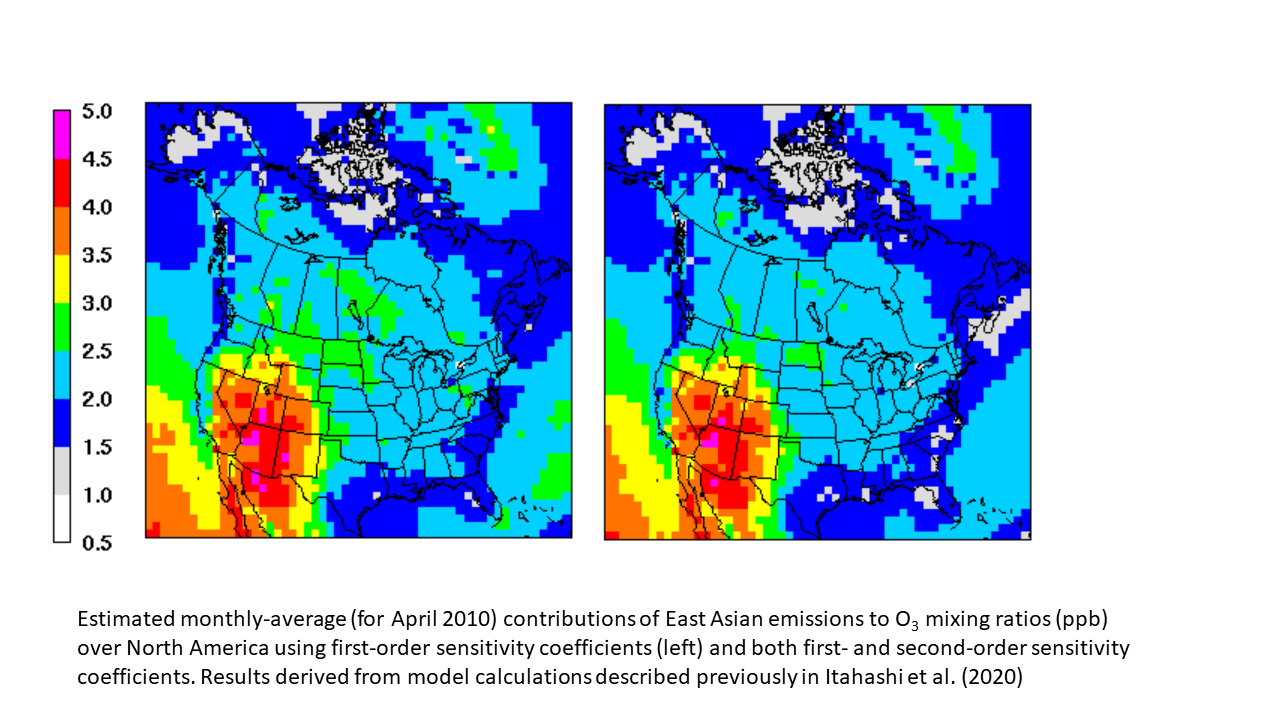
As part of activities of the Air Quality Model Evaluation International Initiative (AQMEII) and Task Force on Hemispheric Transport of Air Pollution (TF-HTAP), Hogrefe et al. (2018) conducted extensive comparisons of three-dimensional tropospheric O3 distributions derived from the HCMAQ model for the year 2010 with those from contemporary global chemistry transport models, including the Chemistry – Integrated Forecasting Model (C-IFS) (Flemming et al., 2015), the Goddard Earth Observing System model coupled to chemistry (GEOS-Chem) (Bey et al., 2001), and AM3 (Donner et al., 2011). Comparisons of time-height cross sections of monthly-mean O3 mixing ratios at locations of several ozonesonde launch sites across North America (Figure 8 in Hogrefe et al., 2018) indicate that HCMAQ model was able to reproduce the seasonal variations in the observed three-dimensional O3 structure both at locations nominally representative of inflow along the western and northern boundaries (at sites Trinidad Head, Edmonton, Churchill) as well as across the contiguous U.S. (at sites Boulder, Huntsville, Wallops Island). Additionally, both the magnitude and the seasonal variations in model bias and error in HCMAQ were similar and in range of those from the other 3 large scale models. Comparison of monthly mean O3 variations at 500mb at the 6 sites (Figure 9 in Hogrefe et al., 2018) further illustrate that the HCMAQ model is able to capture the influence of large-scale flow patterns on tropospheric O3 distributions and that its skill is comparable if not better than the other modeling systems. Extending the analysis presented in Hogrefe et al. (2018), Figure S2 presents a comparison of surface-750mb average O3 distributions derived from HCMAQ with those inferred from measurements at six ozonesonde sites. Also shown in the figure is the range of predictions from the other 3 global models. Note that surface-750mb average is indicative of the “PBL-average” values used in the analysis in the manuscript main text and thus directly representative of the performance of the O3 concentration metric used in the analysis presented in the main text. As illustrated in Figure S2, across all sites, the difference between the HCMAQ predictions and observations is much smaller than the range of predictions from the other models. These comparisons further show that the model replicates the seasonal variations in lower tropospheric O3 across the U.S. with skill comparable to and often better than other contemporary global chemistry-transport models.



**Figure S2**: Comparison of monthly-mean “PBL-average” (surface-~750mb) ozone predictions with ozonesonde measurements at several locations across the U.S. for the year 2010. Blue line represents HCMAQ predictions, while the dash red lines represent the range of predictions (minimum and maximum) from the other global chemistry-transport models analyzed in Hogrefe et al. (2018). Shaded area depicts the range of observations averaged over varying layer structures of the different models.

**S2 Impacts of using only first-order sensitivity coefficients on estimates of LRT O3**

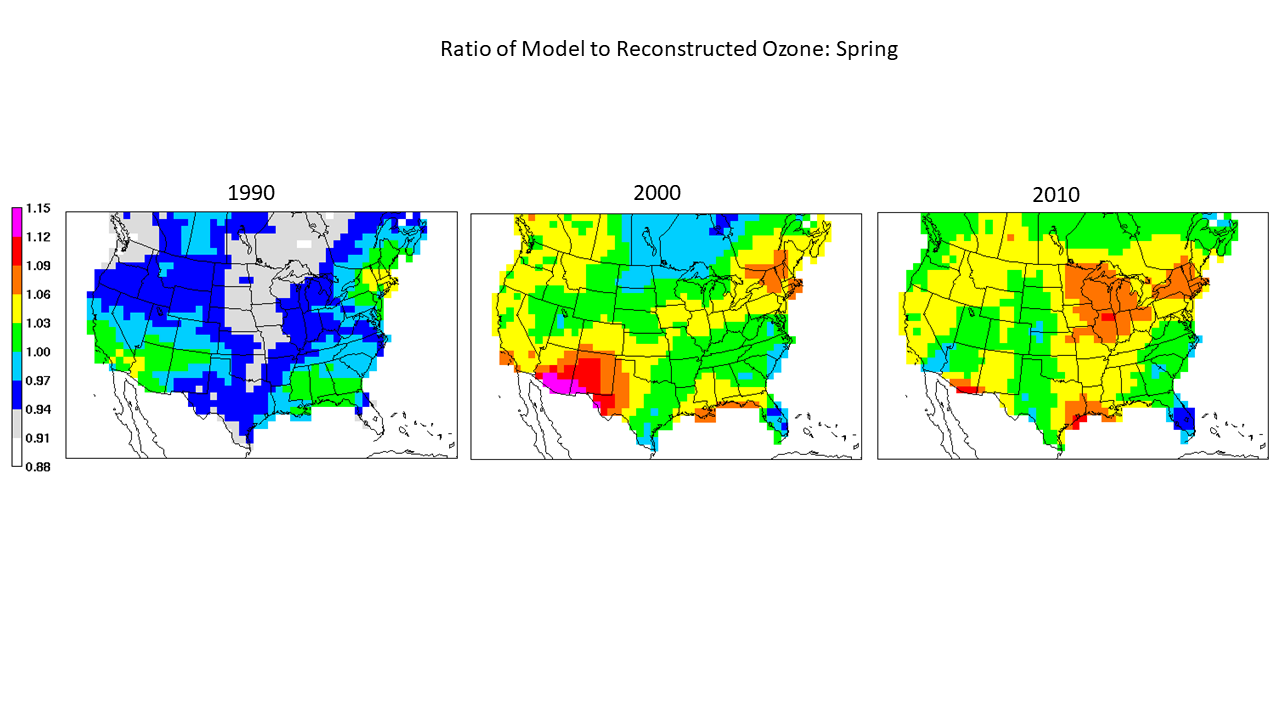
Since O3 production is dependent on both NOx and VOC emissions, calculating both first and second order sensitivity coefficients for O3 to both precursors and for each source region for extended simulation periods becomes computationally prohibitive. Thus, in the current study we tracked only first order sensitivity coefficients of O3 to NOx and VOC emissions from each of the source regions in Figure 1 of the main text. To examine the impact of this assumption and not using source specific higher order sensitivity coefficients, we compared the estimated impacts of East Asia emissions on long range transported O3 across North America using the sensitivity calculations in our previously analysis reported in Itahashi et al. (2020). Figure S3 compares the zero-out contributions of East Asia emissions on monthly mean surface-level O3 mixing ratios across North America using only the first order sensitivity coefficients for East Asia NOx and VOC emissions with estimates using both first and second-order sensitivity coefficients for East Asia NOx and VOC emissions. As illustrated in Figure S3, the estimated far field impacts of East Asia emissions on long-range transported O3 to North America (both magnitude and spatial distribution) are very similar for the two cases, indicating very minimal impact of this approximation on our study results and conclusions related long-range transport from distant emission sources.



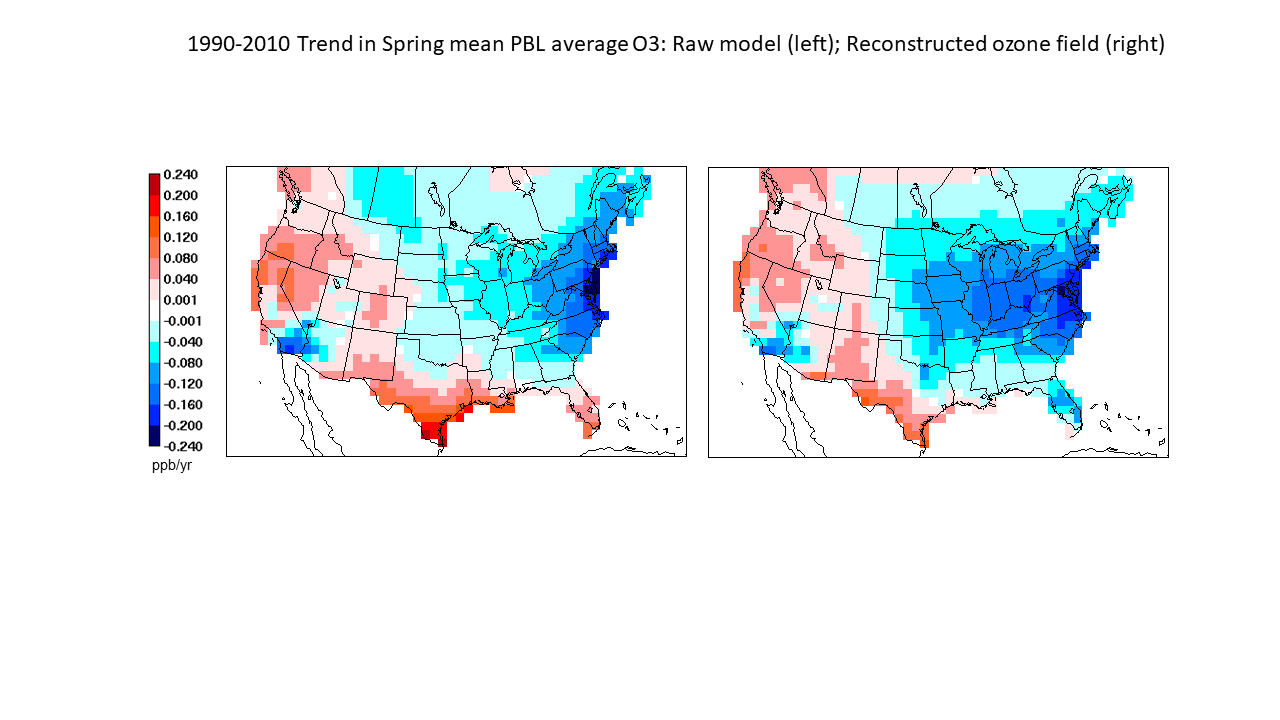
**Figure S3**: Estimated monthly-average (for April 2010) contributions of East Asian emissions to O3 mixing ratios (ppb) over North America using first-order sensitivity coefficients (left) and both first- and second-order sensitivity coefficients. Results derived from model calculations described previously in Itahashi et al. (2020).

**S3 Comparison of reconstructed and original O3 fields**

In order to compare the reconstructed O3 field derived from Equation 12 with the original fields from the model, Figure S4 presents the ratio of the original field to the reconstructed field. These correspond to Spring-mean PBL-average values for three representative years. For a majority of the grid cells across the Contiguous U.S., the reconstructed values based on Equation 12 in the main text are within 6% of the original model estimate and typically within 15%. Comparison of 1990-2010 trends in PBL-average O3 during Spring (Figure S5) further suggest that the trends inferred from the reconstructed fields largely match those inferred from the original field across a majority of the contiguous U.S. and show consistency in the regions with increasing or decreasing trends with the exception in regions of south Florida and portions along the Gulf coast, where the coefficient of determination (R2) for the linear regression estimating the trends are also very low. This may be due to meteorological variability that is not captured adequately even in the original coarse resolution model calculations.



**Figure S4**: Ratio of modeled and reconstructed PBL-average ozone fields during Spring for three representative years: 1990, 2000, and 2010



**Figure S5**: Comparison of 1990-2010 trends in PBL average O3 during Spring based on the model output (left) and the reconstructed field based on Equation 12 in the main text

**Additional Supplemental Figures**

Diagram

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**Figure S6**: Estimated contributions of different source regions to O3 distributions across the Northern Hemisphere for Summer (June-July-August) 2006. (a) PBL average O3 during Summer 2006. Fractional contributions of individual source regions estimated from the first-order sensitivities: (b) NAM, (c) EUR+RUS, (d) NAF+MDE, (e) Other, (f) CAM, (g) EAS+SEA, (h) SAS and (i) stratosphere.

Diagram, timeline

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**Figure S7**: Estimated contributions of different source regions to O3 distributions across the Northern Hemisphere for Winter (December-January-February) 2006. (a) PBL average O3 during Winter 2006. Fractional contributions of individual source regions estimated from the first-order sensitivities: (b) NAM, (c) EUR+RUS, (d) NAF+MDE, (e) Other, (f) CAM, (g) EAS+SEA, (h) SAS and (i) stratosphere.

Diagram

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**Figure S8**: Estimated contributions of different source regions to O3 distributions across the Northern Hemisphere for Fall (September-October-November) 2006. (a) PBL average O3 during Fall 2006. Fractional contributions of individual source regions estimated from the first-order sensitivities: (b) NAM, (c) EUR+RUS, (d) NAF+MDE, (e) Other, (f) CAM, (g) EAS+SEA, (h) SAS and (i) stratosphere.

Map

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**Figure S9**: Estimated imported O3 (defined as amount of O3 transported from the 7 source regions in Figure 1 and due to stratosphere-troposphere exchange) distributions across the United States and surrounding regions for Winter 2006 for (a) the surface to 2km average and (b) the model’s surface-layer.

Diagram

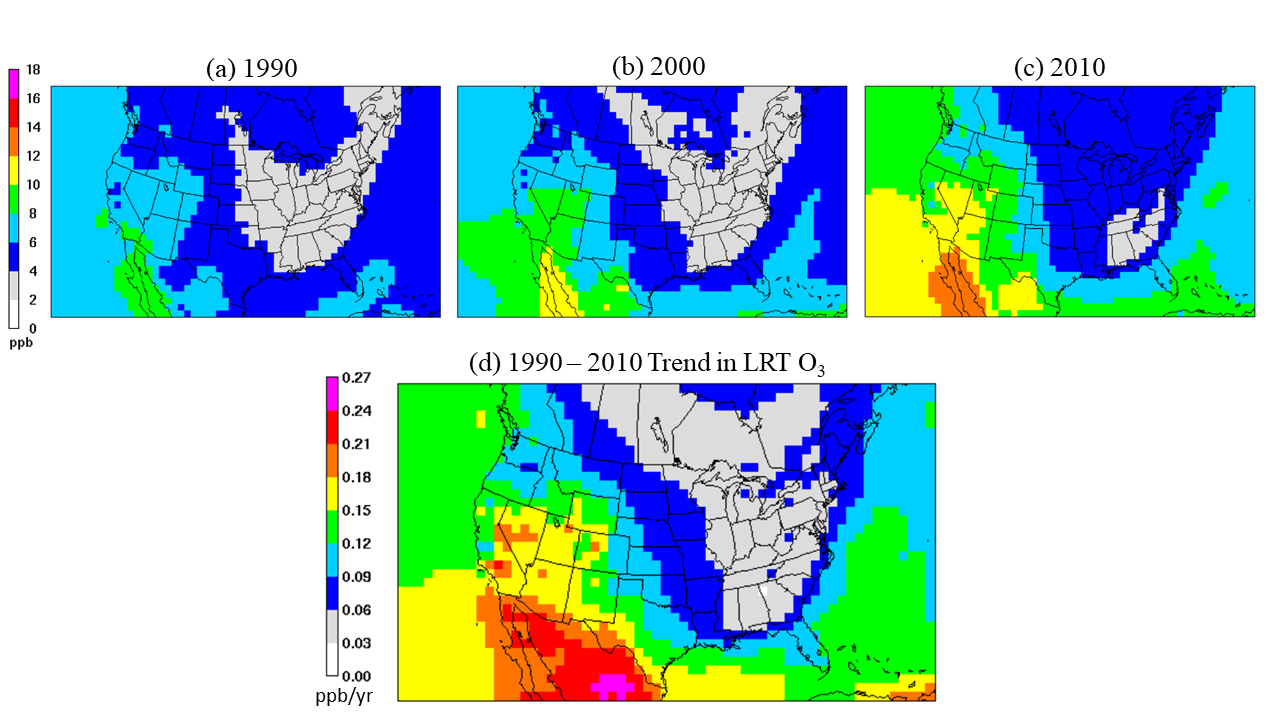
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**Figure S10**: Fractional contributions of the different sources to the 0-2km average Winter 2006 estimated imported O3 mixing ratios in Figure S3.

Diagram

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**Figure S11**: Fractional contributions of the different sources to the Spring 2006 estimated imported O3 mixing ratios at the surface shown in Figure 4c.



**Figure S12**: 1990-2010 changes in long-range transported O3 to North America during Summer. Estimated long-range transported O3 to North America from the different source regions for (a) 1990, (b) 2000, and (c) 2010. The estimated trend in long-range transported O3 to North America for the 1990-2010 period is illustrated in (d). The trend is estimated as the slope of linear regression of the estimated seasonal-mean long-range transported O3 mixing ratios for each of the individual years in the 1990-2010 period. All estimates are for the 0-2km average O3 mixing ratios.

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