Multi-model Estimates of Intercontinental Source-Receptor Relationships for Ozone Pollution

A.M. Fiore[1], F.J. Dentener[2], O. Wild[3], C. Cuvelier[2], M.G. Schultz[4], P. Hess[5], C. Textor[6,7], M. Schulz[7], R. Doherty[8], L.W. Horowitz[1], I.A. MacKenzie[8], M.G. Sanderson[9], D.T. Shindell[10], D.S. Stevenson[8], S.Szopa[7], R.Van Dingenen[2], G. Zeng[11], C. Atherton[12], D. Bergmann[12], I.Bey[13], G. Carmichael[14], B.N. Duncan[15], G. Faluvegi[10], G. Folberth[13], M. Gauss[16], S. Gong[17], D. Hauglustaine[7], T. Holloway[18], I.S.A. Isaksen[16], D.J. Jacob[19], J.E. Jonson[20], J. W. Kaminski[21], T.J. Keating[22], A. Lupu[21], E. Marmer[2], V. Montanaro[23], R. Park[19,24], G. Pitari[23], K.J. Pringle[9,25], J.A. Pyle[11], S. Schroeder[4], M.G.Vivanco[26], P. Wind[20], G. Wojcik[27], S. Wu[19], A. Zuber[28]

1. Geophysical Fluid Dynamics Laboratory, NOAA, Princeton, NJ, USA.
2. European Commission, DG-Joint Research Centre, Institute for Environment and Sustainability, Ispra, Italy.
3. Department of Environmental Science, Lancaster University, UK.
4. ICG-2, Forschungszentrum-Jülich, Germany.
5. National Center for Atmospheric Research, Boulder, CO, USA.
6. Service d'Aéronomie, CNRS/UPMC/IPSL, Paris, France.
7. Laboratoire des Science du Climat et de l'Environment, Gif-sur-Yvette, France.
8. School of GeoSciences, University of Edinburgh, UK.
9. Met Office Hadley Centre, Exeter, UK.
10. NASA Goddard Institute for Space Studies and Columbia University, New York, NY, USA.
11. National Centre for Atmospheric Science, Department of Chemistry, University of Cambridge, UK
12. Atmospheric Science Division, Lawrence Livermore National Laboratory, CA, USA.
13. EPFL, Lausanne, Switzerland.
14. Center for Global and Regional Environmental Research, University of Iowa, Iowa City, IA, USA.
15. Goddard Earth Sciences & Technology Center, UMBC, MD, USA.
16. Department of Geosciences, University of Oslo, Norway.
17. Science and Technology Branch, Environment Canada, Toronto, Canada
18. Center for Sustainability and the Global Environment, University of Wisconsin-Madison, Madison, WI, USA.
19. Atmospheric Chemistry Modeling Group, Harvard University, Cambridge, MA, USA.
20. Norwegian Meteorological Institute, Oslo, Norway.
21. Center for Research in Earth and Space Science, York University, Canada.
22. Office of Policy Analysis and Review, Environmental Protection Agency, Washington DC, USA.
23. Dipartimento di Fisica, Universita de L'Aquila, Italy
24. Now at Seoul National University, Korea.
25. Now at Max Planck Institute for Chemistry, Mainz, Germany.
26. CIEMAT, Madrid, Spain.
27. Northrop Grumman Corporation, VA, USA.

28. Environment Directorate General, European Commission, Brussels, Belgium.

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Abstract

A thorough understanding of the surface O_3 response over a "receptor" region to emission changes over a foreign "source" region is needed to evaluate the potential gains from an international approach to abate ozone (O_3) pollution. We apply an ensemble of 21 global and hemispheric chemical transport models to estimate the average surface O_3 response over East Asia (EA), Europe (EU), North America (NA) and South Asia (SA) to 20% decreases in anthropogenic emissions of the O_3 precursors, NO_x, NMVOC, and CO (individually and combined), from each of these regions. The ensemble mean O_3 matches observed surface concentrations throughout the year over EU but overestimates them by >10 ppb during summer and early fall over NA and EA. The sum of the O_3 responses to NO_x, CO, and NMVOC perturbations is approximately equal to that with all three precursors reduced together, and we focus here on the response to combined reductions. The most robust annual mean responses across the models are that surface O_3 concentrations over EU are most influenced by anthropogenic emissions reductions over NA (ensemble mean: 0.4 ppb; inter-model range: 0.2-0.5 ppb), and that emissions reductions over SA have the smallest effect on the other receptor regions (< 0.2 ppb). Emission reductions in the three foreign regions decrease annual mean surface O_3 over the Asian regions by 0.2-0.4 ppb each, and reductions in EA and EU decrease annual mean O_3 by 0.2 ppb each over NA, although the relative impacts vary by model. Applying these ensemble annual mean results to changes in anthropogenic emissions from 1996 to 2002, we estimate a Northern Hemispheric increase in background surface O_3 of about 0.1 ppb yr^{-1}, at the low end of the 0.1-0.5 ppb yr^{-1} derived from observations. The ensemble mean surface O_3 response to emission reductions in the 3 foreign regions combined is largest in spring and late fall (~0.8 ppb in all regions). We define an "import sensitivity" to gauge the relative contribution of foreign versus "domestic" (*i.e.*, over the source region itself) emission changes for each region. This ratio ranges from 0.5 to 1.1 during the months of maximum foreign influence, and from 0.2 to 0.5 when domestic influence is largest. From an additional simulation in which global atmospheric methane was reduced, we infer that 20% reductions in regional anthropogenic methane emissions would yield an O_3 response over foreign receptor regions roughly equal to that produced by 20% reductions of anthropogenic NO_x, NMVOC and CO emissions.

1. Introduction

Reducing aerosol and ozone (O_3) levels in surface air is desirable for improving public health as exposure to these atmospheric constituents aggravates respiratory illness and may lead to premature mortality [*World Health Organization*, 2005]. Findings from numerous observational and modeling studies indicate that long-range transport of pollutants degrade air quality over remote continents [*e.g.*, *Wilkening et al.*, 2000; *Holloway et al.*, 2003; *Akimoto et al.*, 2003]. Satellite images of aerosols, particularly dust and smoke, illustrate the capacity for dust storms and biomass burning to influence tropospheric composition on a hemispheric scale [*e.g.*, *Husar et al.*, 2001]. Ground-based measurements of aerosol composition provide evidence for a foreign influence in surface air; for example, the presence of smoke from Siberian fires and dust from Asia and Africa over the United States [*e.g.*, *Prospero*, 1999; *Jaffe et al*., 2003a, 2004]. In contrast, attributing O_3 pollution to a specific source region is complicated by the interplay of processes influencing intercontinental transport (export from the source region, non-linear chemical processing and dilution in transit, and mixing with surface air over the receptor region), and by a large hemispheric background and the dominance of local emissions in contributing to high-O_3 events [*e.g.*, *Derwent et al.*, 2003; *Fiore et al.*, 2003; *Goldstein et al*., 2004; *Jonson et al*., 2005]. Given the difficulty of diagnosing O_3 source-receptor (SR) relationships (*i.e.*, the change in O_3 over a receptor region produced by emission changes within a source region) from observations, estimates of these relationships rely heavily on models. Here, we use an ensemble of 21 global and hemispheric chemical transport models (CTMs; Table A1) to quantify the impact of O_3 precursor emissions from four major continental-scale source regions in the Northern Hemisphere on surface O_3 in the same four "receptor" regions (Figure 1). Prior studies indicate that multi-model mean results better represent a range of observations than any individual model [*Schulz et al.*, 2006; *Stevenson et al*., 2006; *Reichler and Kim*, 2008]; the range of results across individual models further provides a measure of uncertainty in our understanding as represented in the current generation of CTMs.

Tropospheric O_3 is produced via the photochemical oxidation of volatile organic compounds (VOC) and carbon monoxide (CO) in the presence of nitrogen oxides (NO_x). To date, regulations to abate surface O_3 pollution address emissions of the traditional O_3 precursors (NO_x, non-methane VOC and CO) which react within hours-to-weeks to produce a "short-term" O_3 response. By altering hydroxyl radical (OH) concentrations, perturbations to emissions of these species affect the major loss pathway for methane (CH_4), the most abundant atmospheric VOC and a major precursor to O_3 in the remote troposphere [*Crutzen*, 1973; *Prather*, 1996; *Daniel and Solomon*, 1998; *Fuglestvedt et al*., 1999; *Derwent et al*., 2001; *Collins et al*., 2002], producing a "long-term" influence on surface O_3. This "long-term" O_3 response follows the time scale and the spatial pattern of the O_3 production from CH_4, and somewhat offsets the O_3 response to perturbations in surface NO_x emissions, while enhancing the response to NMVOC and CO emission changes. [*Wild et al*, 2001; *West et al*., 2007]. Anthropogenic CH_4 emissions have also been shown to contribute directly to O_3 in surface air [*Fiore et al*., 2002a; *Dentener et al.*, 2005; *West et al.*, 2007; *Fiore et al*., 2008]. The contributions to surface O_3 over a receptor region both from CH_4 and from the foreign emissions of the traditional O_3 precursors are generally considered to be part of the "background" O_3 level

(along with natural precursor emissions), and are not generally considered in air pollution mitigation strategies.

Analysis of observations at northern mid-latitudes indicate that background O_3 has been increasing in recent years, although estimates vary, with some revealing little change [*e.g., Vingarzan*, 2004; *TF HTAP*, 2007; *Oltmans et al.*, 2006; *Derwent et al*., 2007; *Schultz et al*., 2007]. Several modeling studies suggest that projected increases in emissions around the globe will enhance hemispheric background O_3 in the coming decades, potentially offsetting efforts to improve regional air quality via controls on domestic precursor emissions [e.g. *Jacob et al*., 1999; *Yienger et al.*, 2000; *Collins et al*., 2000; *Fiore et al.* 2002a; *Dentener et al*., 2005; *Derwent et al*., 2006; *Szopa et al.*, 2006; *Ellingsen et al*., 2008]. Efforts to improve air quality typically focus on controlling local and regional sources. In nations where O_3 precursors have been regulated for decades, the combination of increasing hemispheric background levels and mounting control costs could make pursuing international cooperation an attractive option [*Keating et al*., 2004; *Bergin et al*., 2005; *Solberg et al*., 2005]. An international approach to air quality management will require a strong scientific understanding of the SR relationships between continents and nations.

Under the United Nations Economic Commission for Europe (UNECE) Convention on Long Range Transboundary Air Pollution (CLRTAP), the Task Force on Hemispheric Transport of Air Pollution (TF HTAP; www.htap.org) was established to advance the understanding of hemispheric transport of air pollutants in the Northern Hemisphere. A major TF HTAP activity is to coordinate a multi-model effort to quantify and estimate uncertainties in intercontinental SR relationships for O_3, aerosols, mercury, and persistent organic pollutants. Companion manuscripts investigate NO_y deposition [*Sanderson et al*., 2008], transport with idealized tracers [*Schultz et al*, 2008, in prep], the Arctic as a receptor region [*Shindell et al*, 2008], and aerosols [*Schulz et al*, 2008, in prep].

Prior estimates for intercontinental SR relationships differ by factors of about 2 to 6 for a given SR pair among source regions at northern mid-latitudes [TF HTAP, 2007 and references therein]. Comparison among prior studies, however, is limited by methodological differences, including definitions of source and receptor regions, reported metrics, time period of analysis, and SR calculation method [TF HTAP, 2007]. Observational analyses have compared concentration differences in air masses originating from a source region versus a background value [*e.g. Huntrieser et al*., 2005; *Derwent et al*., 1998; *Jaffe et al*., 2003b]. A suite of methods for source attribution have been applied in models, including marking tracers by region of O_3 production [*e.g. Jaeglé et al*., 2003; *Derwent et al*., 2004], labeling by the regional NO_x source contributing to O_3 production [*Hess and Lamarque*, 2007], and sensitivity simulations in which regional emissions are perturbed [*e.g. Jacob et al.*, 1999; *Yienger et al.*, 2000; *Wild and Akimoto*, 2001; *Fiore et al.*, 2002b, *Auvray and Bey*, 2005].

The approach adopted here builds upon these previous studies by applying a consistent experimental design across multiple models to provide an estimate of SR relationships throughout the year. Specifically, we investigate the changes in surface O_3 resulting from fixed percentage reductions in anthropogenic O_3 precursors (NO_x, CO, CH_4, and NMVOC). We first describe the modeling framework (Section 2) and evaluate the base case simulations with observations of surface O_3 (Section 3). Our analysis

focuses on two SR metrics: (1) the response, defined as the spatially averaged absolute change in O_3 concentrations over a receptor region due to emission changes in the source region, and (2) the "import sensitivity", defined as the ratio of the sum of the changes in surface O_3 resulting from perturbations to precursor emissions in the three foreign source regions to the surface O_3 change resulting from the same percentage perturbation to domestic emissions (*i.e.*, emissions within the continental receptor region; Section 4). We then examine the response of surface O_3 to changes in CH_4 levels (Section 5). In an effort to relate our results more directly to statistics commonly employed in air quality management, we also analyze the response of a threshold indicator of air quality to emission changes (Section 6). Finally, we assess the consistency of observed trends in surface O_3 in recent decades to those derived by applying our SR relationships to estimated trends in regional precursor emissions (Section 7).

2. Model Simulations and Emissions

The 21 chemical transport models (CTMs) that conducted simulations designed to estimate source-receptor (SR) relationships for O_3 are described in Table A1 (auxiliary material). Initial results were reported by *TF HTAP* [2007]; we expand here upon that analysis. All modeling groups used meteorology from the year 2001 and horizontal resolution of 5°x5° or finer. Most models were driven by meteorological fields from one of several reanalysis centers (either prescribed directly or via linear relaxation) although 2 models were general circulation models with meteorology generated based on observed sea surface temperatures for 2001 (Table A1). About half of the models had resolutions of 3ºx3º or finer, similar to the ACCENT/AR4 intercomparison [*Dentener et al*, 2006; *Stevenson et al*. 2006]. CH_4 concentrations were set to a uniform mixing ratio of 1760 ppb and modeling groups were requested to use their best estimate for emissions of O_3 precursors (Tables A2 and A3), so the results include uncertainties associated with the use of different emission inventories.

We consider four major source regions at northern mid-latitudes (Figure 1): East Asia (EA), Europe and northern Africa (EU), North America (NA), and South Asia (SA). The EU and EA regions span roughly equivalent areas ($2.1x10^7$ and $2.3x10^7$ km^2), with NA somewhat larger ($2.6x10^7$ km^2) and nearly twice the area of SA ($1.5x10^7$ km^2). According to the model ensemble mean (μ), similar quantities of anthropogenic NO_x and NMVOC are emitted from EU, NA, and EA (to within ~30%; Table 1; see Tables A2 and A3 for the emissions used by individual models). The standard deviation (σ) indicates the diversity of the emission inventories used in the models, with the smallest inter-model spread for anthropogenic NO_x emissions in EU and NA (σ/μ <10%) and the largest spread for anthropogenic VOC from EU (σ/μ = 58%). A comparison of the model ensemble mean anthropogenic and total (which also includes biomass burning and biogenic contributions) emissions in Table 1 shows a dominant contribution (>70%) from anthropogenic NO_x and CO in all regions considered. For the ensemble mean NMVOC emissions, biogenic emissions dominate in all regions except for EU where anthropogenic and biogenic contributions are approximately equal.

In the base simulation (SR1), the 21-model annual spatial mean surface O_3 mixing ratios and their standard deviations over the four continental source regions in Figure 1 are similar: 36.2±3.9 ppb for NA, 37.8±4.5 for EU, 35.8±3.0 for EA, and 39.6±4.0 for

SA. The largest ensemble mean seasonal amplitude (difference between the maximum and minimum months) occurs in EU (19.8±5.9 ppb), possibly reflecting NO_x titration in the stronger wintertime boundary layer compared to the other regions (10.4±2.6 for NA; 12.7±3.2 for EA, and 14.8±6.0 for SA).

Relative to SR1, we conduct 16 sensitivity simulations in which anthropogenic emissions of the traditional O_3 precursors (NO_x, NMVOC, and CO) are reduced by 20% individually (simulations SR3, SR4, and SR5, respectively) and jointly along with aerosols ("ALL"; simulation SR6) within each of the four source regions in Figure 1. These simulations are labeled hereafter according to the respective emission scenario and the region in which emission reductions were applied (*e.g.*, SR3EA identifies the simulations with 20% reductions of anthropogenic NO_x emissions within East Asia). An additional sensitivity simulation was conducted in which the CH_4 mixing ratio was decreased by 20% (to 1408 ppb) and other O_3 precursor emissions were held constant (SR2). All simulations were conducted for a full year, following a minimum of six months initialization, a sufficient time for the simulated trace gas concentrations to fully respond to the imposed emission or concentration perturbations given our use of uniform CH_4 mixing ratios; responses on longer time scales are diagnosed in Section 5.

The perturbation magnitude of 20% was chosen for two reasons: (1) A reduction of this magnitude is expected to be feasible on a decadal time frame and is thereby relevant to air pollution policy, and (2) it reflects a compromise between producing a clear signal in the O_3 simulations and applying a sufficiently small perturbation to allow the results to be scaled linearly to different size perturbations. Under the ACCENT/AR4 Experiment 2, *Stevenson et al.* [2006] found a broadly linear relationship between the 26-model mean tropospheric O_3 burden and global NO_x emissions within the ±50% range of present day emissions considered in that study, although those simulations did not exclusively change NO_x emissions. The scalability of our results to perturbations of other magnitudes is examined further for NO_x in Section 4.2, and has been shown to hold for the O_3 response to changes in CH_4 over the range of present-day anthropogenic emissions [*Fiore et al*., 2008]. We further assume that the O_3 responses to regional perturbations can be summed to approximate the O_3 response to simultaneous reductions in multiple regions.

3. Model Evaluation with Surface Observations

A major challenge to assessing model skill at representing intercontinental SR relationships arises from the difficulty of directly observing these relationships (particularly in surface air). Testing the models with simultaneous measurements of O_3 and related species is preferable [e.g. *Sillman*, 1999]; such observations are mainly limited to intensive field campaigns, which are the focus of an ongoing TF HTAP multi-model study. A companion study will evaluate the models with the ozonesonde network [*Jonson et al*., in prep]. Many of the models in our study have been compared with ozone observations for the year 2000 as part of the ACCENT/AR4 Experiment 2 study [e.g., *Dentener et al*., 2006; *Stevenson et al*., 2006; *Ellingsen et al.*, 2008]. Comparison with observations from the global ozonesonde network [*Logan*, 1999] across different latitude and altitude bands showed that the overall multi-model root mean square error (RMSE) was 5.9 ppb, ranging from 5.1 to 18 ppb across the models [*Stevenson et al.,* 2006].

Annual mean surface O_3 concentrations were within 5 ppb of the measurements in the United States, China, and Central Europe [*Dentener et al*., 2006]. Overestimates of 10-15 ppb were found in Africa, India, and the Mediterranean, for reasons not yet understood [*Dentener et al*., 2006; *Ellingsen et al*., 2008].

Due to limited availability of surface O_3 measurements over India, China, and Africa, we focus here on the widespread observational networks in the United States, Europe, and Japan (Figure 2). While the individual models often differ by more than 15 ppb, the model ensemble mean generally captures the observed seasonal cycle and is close to the observed regional mean. A wide range of simulated tropospheric O_3 budgets has been documented in the literature, attributed in part to factors that are likely to contribute to inter-model variability in simulated surface O_3 concentrations, such as differences in surface emissions of NO_x and isoprene, as well as in model treatment of dry deposition, heterogeneous chemistry and the organic nitrates from isoprene [*Stevenson et al*., 2006; *Wu et al*., 2006; *Wild*, 2007; *Ellingsen et al*., 2008].

We separate the observational sites at low elevations from those at higher altitudes since high altitude sites more frequently sample free tropospheric air and thus are better suited to detecting hemispheric pollutant transport (which occurs most efficiently in the free troposphere) prior to mixing with local pollutant signals in the planetary boundary layer [*Cooper and Moody*, 2000; *Trickl et al*., 2003; *Weiss-Penzias et al*., 2006]. At the high altitude sites (Figures 2c and 2h), the models tend to underestimate O_3 concentrations. Steep topographic gradients that are averaged out within one model grid cell, particularly over Europe, may be responsible if the measurements are more representative of the free troposphere than the models' surface layer (where O_3 deposition leads to lower concentrations). During the cold season, the underestimate could also reflect insufficient transport from the stratosphere [*Shindell et al.*, 2006].

The model ensemble mean and median exhibit little bias at low-altitude European sites and capture the seasonal cycle (Figures 2a and 2b), an apparent improvement over the underestimate in summer months found by *Ellingsen et al*. [2008]. In contrast, the multi-model mean overestimates the observed summertime surface O_3 concentrations over Japan (bias of 12 ppb; Figure 2i) and in the eastern U.S. (bias greater than 15 ppb in July in Figures 2d, 2f and 2g). The observed summer minimum in O_3 over Japan occurs during the wet season of the Asian monsoon. Results from the MICS-Asia regional model intercomparison suggest that the positive model bias may stem from inadequate representation of southwesterly inflow of clean marine air [*Han et al*., 2008; *Holloway et al*., 2008]. Examination of inter-model differences in this region with the TF HTAP idealized tracer transport simulations [*Schultz et al*., in prep] should provide further insights into the source of this problem.

Ellingsen et al. [2008] also found an overestimate of surface O_3 levels in July through September for the year 2000 over the Great Lakes, and in June through September over the Southeastern United States. In July and August, however, the ensemble median value fell within the standard deviation of the observations. We find that the observed July and August average O_3 decreased by 10 ppb from 2000 to 2001; the larger model error shown in Figure 2 than found by *Ellingsen et al*. [2008] suggests that the model ensemble mean does not capture the observed interannual variability over the eastern United States, although our use of different emission inventories from

Ellingsen et al. [2008] may also play a role. The bias is particularly large over the southeastern U.S. where uncertainties in isoprene-NO_x-O_3 chemistry may contribute; smaller biogenic NMVOC emissions over Europe (difference between total and anthropogenic NMVOC in Table 1) would lessen the impact of any problems in this chemistry on surface O_3 concentrations there. The bias is not driven by nighttime processes since restricting our comparison to afternoon hours does not yield any improvement (Figure A1). The models do not account for the observed decrease in U.S. power plant NO_x emission reductions that occurred between 1999 and 2003 (~50% decrease averaged over 53 power plants) [*Frost et al*., 2006]. Comparison with the SR3NA simulations (in which North American anthropogenic NO_x emissions are decreased by 20%, roughly the total emission reduction contributed by the power plants), however, decreases the bias by only 2-4 ppb. The occurrence of the eastern U.S. bias in summer strongly suggests an overestimate of regionally produced O_3. The sensitivity of surface O_3 over all regions to foreign sources is strongest in spring and late autumn (Section 4), however, when the model ensemble mean matches the observed values in all regions, lending confidence to our results for those seasons.

4. Source-Receptor Relationships for NO_x, NMVOC and CO

4.1 Model Ensemble Mean Results

Figure 3 (and Table A4) shows the annual average surface O_3 response in the receptor regions to 20% regional reductions of anthropogenic NO_x, CO, NMVOC emissions, individually and all together ("ALL"), as well as the sum of the responses to emission perturbations in the "3 foreign" source regions. In most receptor regions, O_3 responds most strongly to NO_x, followed by NMVOC and CO, respectively. An exception occurs for EU emissions where the model ensemble O_3 response to NMVOC, for foreign SR pairs, is comparable to that from NO_x. The relative dominance of NO_x diminishes when the long-term feedback through CH_4 is taken into account (Section 5). For all SR pairs, domestic emission reductions are most effective at reducing surface O_3. Surface O_3 also decreases when emissions are reduced in a foreign source region, often by more than 10% of the decrease attained with the same percentage emission reduction applied domestically (Figure 3 and bold entries in Table A4). The largest annual mean responses to foreign emissions are ~50% of the response to domestic emissions and occur for NA NO_x, CO, and ALL on EU surface O_3, and for EU NMVOC on SA surface O_3.

We next examine seasonality in these SR relationships, beginning with the seasonal cycle in the O_3 response to domestic emission reductions (filled circles in Figure 4). Over all regions, the domestic response to the 20% decrease in CO emissions (~0.1 ppb) varies little during the year while the response to NO_x exhibits the strongest seasonality (max of >1 ppb). Over EU, NO_x reductions increase the model ensemble mean O_3 from November to March. The response to NMVOC emission reductions is largest in boreal winter (up to ~0.5 ppb over EU, though seasonality is weak), when biogenic emissions, radiation, and humidity are at their seasonal minimum and O_3 production is more sensitive to anthropogenic NMVOC [*Jacob et al*., 1995]. The seasonality of the domestic response to ALL is largely driven by NO_x, peaking in summer for NA, EU, and EA, and in October through March for SA. The different

seasonality over SA reflects the influence of the Asian monsoon (wet season during summer).

We find a larger contribution of intercontinental transport at northern mid-latitudes to surface O_3 concentrations during boreal spring and fall (filled circles in Figure 5), reflecting a combination of more frequent storm tracks that enhance ventilation of the continental boundary layer, more efficient transport in stronger mid-latitude westerly flow in the free troposphere, and a longer O_3 lifetime allowing for a longer transport distance than in summer when O_3 chemical production and loss are largest [*Wang et al*., 1998; *Jaffe et al*., 1999; *Yienger et al*., 2000; *Bey et al*., 2001;*Wild and Akimoto*, 2001; *Liu et al*., 2003; *Stohl et al*., 2002; *Weiss-Penzias et al*., 2004; *Wild et al*., 2004; *Holzer et al*., 2005; *Holloway et al*., 2008]. Over all regions, the response to 20% decreases in anthropogenic NO_x emissions from all 3 foreign regions combined are largest in boreal spring and fall to early winter (up to ~0.4 ppb); the response to foreign NMVOC emissions is largest in winter through early spring (0.2-0.4 ppb). Over NA, the model ensemble average O_3 response to foreign NO_x and NMVOC are similar (~0.3 ppb) in winter, spring and summer, although this result varies across models (Section 4.3). The 20% reductions in foreign emissions of CO have little influence (<0.2 ppb) on surface O_3 over the receptor regions, but this influence increases when the long-term feedback via CH_4 is included (Section 5.2).

In Figure 6, we show the surface O_3 response in each receptor region to ALL, and decompose the total foreign impact into the contributions from each of the three foreign source regions. For the NA receptor region, EA and EU contribute similarly from April through November. NA contributes most strongly to EU throughout the year, reflecting its relative upwind proximity. The SA source region exerts a minor influence throughout the year over NA and EU (always less than 0.1 ppb to surface O_3), as SA pollution is typically funneled away from the northern mid-latitude westerlies (the dominant transport pathway to those regions) [*e.g., Li et al*., 2001; *Lelieveld et al*., 2002a; *TF HTAP* 2007], and tends to remain isolated from mid-latitude air [*Bowman and Carrie*, 2002; *Hess*, 2005]. Over EA, emissions from the three foreign source regions induce a similar response in surface O_3 during summer, with stronger sensitivity to emissions in EU (followed by NA) during spring, and to NA (followed by EU) in winter. Over SA, the surface O_3 response is largest when emissions are reduced in EU, except for the November peak which is driven by emissions from EA. We conclude that EU is influenced most strongly by NA throughout the year, but that the other receptor regions exhibit similar responses to emission reductions in at least two of the foreign source regions.

In order to compare the surface O_3 response to foreign versus domestic emissions, we define an "import sensitivity" (IS_r):

$$IS_r = \left(\sum_{f=1}^{3} \Delta O3_{fr} \right) / \Delta O3_{rr}$$

where IS_r represents the import sensitivity for receptor region r; $\Delta O3_{fr}$ represents the model ensemble mean change in surface O_3, spatially averaged over the receptor region r, produced by a 20% decrease in anthropogenic emissions over the foreign source region (f); $\Delta O3_{rr}$ is the change in surface O_3 resulting from the 20% reduction of anthropogenic emissions within the domestic source region. The larger the value of the import sensitivity, the greater the influence of foreign emissions. The annual mean IS_r ranges

from approximately 0.3 (NA and SA) to 0.75 (EU) for NO_x alone; 0.4 (EU) to 1.1 (SA) for NMVOC alone; and 0.45 (NA and SA) to 0.75 (EU) for the combined reductions in all O_3 precursors. The IS_r is typically largest for CO, ranging from approximately 0.9 (EA) to 1.1 (EU), reflecting the longer CO lifetime and the correspondingly smaller influence from domestic sources.

Monthly mean IS_r estimates from the ALL simulations are shown in Figure 7. Over EA, NA, and EU, the IS_r exceed one during boreal winter (also in spring and fall over EU) when O_3 production from domestic emissions is small (Figure 4). Even in summer, when domestic O_3 production peaks, IS_r is 0.2-0.3 over these regions. During the month with the largest surface O_3 response to ALL foreign emission reductions (Figures 5 and 6), IS_{NA}= 0.6 (April), IS_{EU}=0.7 (April), and IS_{EA}=1.1 (March). The IS_{SA} for ALL varies little during the year and is ~0.5 in November and during the broad secondary peak from January through April (Figures 6 and 7). We conclude that the O_3 response to foreign emissions is not negligible when compared to the response to domestic emissions, and is particularly strong in spring and fall at northern mid-latitudes.

4.2 Applicability of Results to Other Emission Perturbations

Evaluating whether the O_3 response to multi-component emission reductions is equivalent to the sum of the responses to single component emission reductions is critical for determining the applicability of our results to other combinations of precursor emission reductions. In order to assess the additivity of our simulations, we construct the ratio of the sum of the O_3 response to 20% reductions in emissions of the individual precursors (SR3-SR1)+(SR4-SR1)+(SR5-SR1) to the O_3 response in the simulation where all precursors (along with aerosols) were reduced simultaneously (SR6-SR1). With one exception, models including reductions of aerosols and precursors in SR6 indicate that the sum of the responses to single component emission reductions exceeds that to multi-component perturbations, by as much as 50% for some SR pairs (Table A5 and Figure A3). Ranking all of the models by the NA domestic response shows that the degree of additivity varies by region within individual models (Figure A3). In Figure 8, we restrict our analysis to those models without aerosol changes in SR6, and find that the responses to single-component versus multi-component emission reductions are approximately equivalent for the O_3 responses to domestic and foreign emissions. We conclude that combined reductions in emissions of aerosols and O_3 precursors dampen the O_3 response relative to that produced by emission reductions of the O_3 precursors alone.

If we wish to apply the responses diagnosed in Section 4.1 more broadly, we need to determine if the O_3 response is sufficiently linear to yield accurate results when scaling to emission perturbations of other magnitudes. To address this point, additional simulations with varying sizes of perturbations to EU anthropogenic NO_x emissions were conducted in the FRSGC/UCI model [*Wild et al*., 2003]. The FRSGC/UCI model tends to produce a larger surface O_3 response to NO_x than the model ensemble mean, but is not an outlier in any month, so we expect these results to apply generally to our model ensemble. Figure 9 shows the response of surface O_3 over EU and NA. The EU source region was chosen as it exhibits the most non-linear response of all regions (*e.g.*, reversing sign with season in Figure 4). The response over the source region (EU) deviates more from that obtained by a linear scaling of the response in the 20%

perturbation simulation than over a remote receptor region (NA). For example, the summertime response over the EU remains linear only for perturbations within 20%, whereas it is linear well beyond perturbations of 50% over NA. The scalability of the results shows that the response is most linear in summer over the foreign receptor region. A companion manuscript [*Wu et al.*, in preparation] will explore the processes contributing to the seasonal dependence in Figure 9, incorporating simulations from other models, perturbations to other O_3 precursor emissions and emissions from other source regions.

4.3 *Robustness of Results as Measured by Inter-model Differences*

It is notoriously difficult to attribute uncertainty to model results. The range spanned by the SR relationships among the models provides a measure of uncertainty as encompassed by differences in the emission inventories and the representations of transport and photochemical processes in the individual models. The uncertainty (measured by the standard deviation divided by the mean (*i.e.*, σ/μ)) associated with the O_3 response to decreases in NMVOC emissions is often larger than that due to reductions in either NO_x or CO emissions (Table A4), and probably reflects the larger uncertainty associated with the NMVOC inventories and the incorporation of the individual NMVOC species into the model chemical mechanisms (Table 1).

Figure 5 suggests that surface O_3 over NA has a similar sensitivity to foreign emissions of NO_x and NMVOC, but this result varies across the individual models (not shown). For example, Figure 10 shows that the magnitude of the surface O_3 response over NA to decreases in EU anthropogenic NMVOC emissions in the individual models correlates ($r^2 = 0.50$) with the anthropogenic EU NMVOC emission total, which varies by nearly a factor of 10. Better constraints on the total NMVOC emissions and their partitioning into NMVOC species with different reactivity should help to reduce the associated uncertainty in the O_3 sensitivity. While the model ensemble mean response in Figure 5 is most sensitive to NO_x, this result only holds for all individual models over SA. We recommend a future study to probe more deeply into the sources of model differences in the surface O_3 sensitivity to NO_x versus NMVOC.

We examine whether the bias in the individual models over the eastern United States (Section 3) manifests as a larger response, for example, of EU surface O_3 to the reductions in NA emissions. We find little correlation with the simulated SR relationships ($r^2<0.1$ for the EU response to NA emission changes in July). The source of the bias, however, should be understood to increase confidence in the estimates for summertime SR relationships for the NA and EA regions.

For each region, we further assess the robustness of the SR relationships in Figure 6 across the 15 individual models. We focus here on the springtime (March, April and May) and annual average response to the combined emissions reductions (*i.e.*, SR6-SR1). The rankings for the EU receptor region are most robust, with NA>EA>SA in all models except for one where the annual mean response to emissions in EA and SA are equal. All models also indicate that annual and springtime mean O_3 over EA, EU, and NA is less sensitive to emissions from SA versus the other three foreign regions. The model spread in the response of NA surface O_3 to emission reductions in EU versus EA in spring indicates more uncertainty, with 10 models predicting equivalent responses (to within 20%), but in 4 models the response to EA emissions is 30-55% greater than that to EU

emissions. For the annual mean NA O_3 response, 8 models predict equivalent responses while 6 suggest that the response to EA emissions is 25-50% larger than that to EU emissions. Over EA, where the winds are northwesterly during spring, the O_3 response to emission changes in EU and NA exceeds that to SA (by a factor of 1.6-3 for EU in 12 models, and 1.5-2 for NA in 9 models); even in the annual mean results, 12 models simulate a larger response to emissions from EU and/or NA than from SA. Over SA, the O_3 response to emission changes rank as EU>NA>EA in 9 of 15 models for spring (but this ranking holds for only 3 models in the annual mean, with 9 models predicting annual mean responses within 20% in at least two of the source regions).

The rankings of individual models often change with season, likely reflecting seasonal variations in transport pathways. Year-to-year fluctuations in transport pathways may also alter the rankings of the foreign source influence. For example, the NA influence on EU surface O_3 in late winter and early spring in years with a positive Arctic Oscillation (AO) index is likely larger than that estimated for 2001 (negative AO) since a positive AO index is associated with warm conveyor belts positioned to transport pollution more efficiently from the northeastern United States to EU [*Eckhardt et al*., 2004; *Lamarque and Hess*, 2004; *Hess and Lamarque*, 2007]. The influence of EU emissions on NA surface O_3 is also expected to be stronger in years with a positive AO index [*Hess and Lamarque*, 2007]. With respect to indices for trans-Pacific transport, the year 2001 should be representative of an average year [*Liang et al*., 2005; *Liu et al*., 2005]. The robustness of the rankings identified here to interannual variability deserves further study.

4.4 Comparison with prior estimates of intercontinental source-receptor relationships

In Figure 11, we compare our results for the NA, EU, and EA regions with the studies referenced in the TF HTAP [2007] that report annual and seasonal mean SR relationships, supplemented by recent analyses by *Lin et al.* [2008], *Duncan et al.* [2008], and *Holloway et al*. [2008]. While the range across previous studies reflects a variety of regional definitions, reported metrics, meteorological years, and methods for source attribution, the consistent modeling approach adopted here restricts the uncertainty as represented by the range across our model results to differences in emissions, chemistry, and transport. Information regarding import to or export from the SA region is limited; our results in Figure 5 for SA are consistent with those of *Kuhnikrishnan et al*. [2006] in showing an autumn peak, but in contrast to that study, we find that the response of SA surface O_3 to EA NO_x emissions is larger in winter than in summer.

In cases where an emission perturbation was not a 100% decrease, we scale the reported O_3 response to estimate the total contribution from the foreign emissions. For example, we use 5*(SR1-SR6) to estimate the total contribution of anthropogenic emissions of the traditional O_3 precursors (NO_x+NMVOC+CO) from a foreign source to a receptor region. Figure 10 implies that a linear scaling of our 20% reductions in anthropogenic NO_x emissions will yield a smaller response than that in a simulation where emissions are set to zero. We expect the response to the combined NO_x+NMVOC+CO emission reductions to deviate less from linearity due to the competing effects on OH from NO_x versus NMVOC+CO but this assumption needs further investigation. The range in annual mean SR relationships for the EU and EA receptor regions across our model ensemble narrows considerably from the estimates in

the literature, with the model ensemble response in Figure 11 smaller than most prior estimates for spring and summer. Our study provides a comprehensive view of the seasonality of SR relationships and a previously unavailable quantitative measure of uncertainty in these relationships.

5. Contribution from CH_4 to the long term response of O_3

5.1 Surface O_3 response to CH_4 concentrations

The annual mean surface O_3 response to a 20% decrease in global CH_4 concentrations is 1.1-1.3 ppb over the receptor regions, largest in SA and EU, followed by NA and EA (Table A4). The O_3 responses in the individual models, however, differ by ~1 ppb. This uncertainty is likely to stem from model differences in OH and NO_x distributions [*e.g., Fiore et al*., 2008]. As the O_3 response to CH_4 is approximately linear over the range of present-day anthropogenic emissions [*Fiore et al*., 2008], scaling the ensemble mean responses in Table A4 from 20% to 100% implies that CH_4 presently contributes 5.5-6.5 ppb of the surface O_3 in the receptor regions.

The tropical location of SA leads to the stronger O_3 response to CH_4 since CH_4 oxidation predominantly occurs in the tropical lower troposphere due to the combination of OH availability and the temperature sensitivity of the CH_4-OH rate constant [*e.g.*, *Spivakovsky et al*., 2000]. One might expect that EU, situated at more northerly latitudes than the other regions, would exhibit the weakest response of surface O_3 to CH_4. Instead, the response is nearly as strong as that found in SA, and stronger than the responses in NA and EA. The largest seasonal amplitude of the O_3 response to CH_4 (difference between the maximum and minimum monthly mean values) occurs in EU, followed by SA, NA, and EA (Figure A4). The O_3 response to CH_4 over EU is largest during summer, possibly reflecting a combination of the stronger seasonality in O_3 production in this northernmost region and stronger CH_4-sensitivity arising from smaller biogenic (high reactivity) VOC emissions than in the other regions. The seasonal amplitude of the EU O_3 response to CH_4 in the individual models is indeed somewhat correlated with the ratio of EU anthropogenic to total NMVOC emissions (r = 0.6 for 13 models).

5.2 Long-term O_3 response to NOx, CO, NMVOC emissions reductions

Perturbations to NO_x, NMVOC and CO emissions influence the oxidizing capacity of the atmosphere (OH), which can change the CH_4 lifetime and thereby contribute a "long-term" change in tropospheric O_3 on the decadal time scale of the CH_4 perturbation lifetime [*e.g. Prather*, 1996; *Daniel and Solomon*, 1998; *Fuglestvedt et al*., 1999; *Wild and Prather*, 2000; *Derwent et al*., 2001; *Wild et al*., 2001; *Collins et al*., 2002; *Stevenson et al.*, 2004]. *West et al*. [2007] previously found that the long-term impacts of 20% decreases in global anthropogenic emissions on population-weighted average surface O_3 at northern mid-latitudes enhanced the short-term response by 16-21% for CO, and decreased it by 6-14% for NO_x, with little long-term influence from NMVOC. This long-term impact on surface O_3 exhibits the spatial distribution of the surface O_3 response to changes in CH_4, which we obtain from the difference in O_3 between the SR2 and SR1 simulations (Section 5.1). By setting atmospheric CH_4 to a uniform, fixed value of 1760 ppb, the SR3 through SR6 simulations neglect the feedback on CH_4 from the changes in OH induced by the 20% decreases in regional anthropogenic

NO_x, NMVOC, and CO emissions. In order to account for this feedback, we first estimate what the CH_4 concentration change would be in a simulation with the same CH_4 loss by OH as in the perturbation simulation, but with CH_4 emissions held equal to those implied by the 1760 ppb atmospheric abundance in SR1.

We apply the formulation of *West et al*. [2007] to estimate the CH_4 abundance that would result from the changes in the other O_3 precursor emissions:

$$[CH_4]_{SRNxx} = [CH_4]_{SR1} * (\tau_{SRNxx}/ \tau_{SR1})^F$$

where SRN represents SR3 through SR6; xx is the 2-letter regional abbreviation in Figure 1; τ_{SR1} is the total atmospheric CH_4 lifetime in the base simulation; τ_{SRNxx} is the CH_4 lifetime in the perturbation simulation; and F is defined as the ratio of the atmospheric response (perturbation) time to the global atmospheric lifetime (see below). Table 2 lists the subset of models that archived the CH_4 loss rates required to determine τ_{SR1}, τ_{SRNxx}, and F. The model ensemble mean $\tau_{SR1} = 8.55 \pm 1.6$ is within 2% of the 26-model mean of 8.67 ± 1.32 reported by *Stevenson et al.* [2006]. From the SR1 and SR2 simulations, we calculate F following *Wild and Prather* [2000]:

$$F = 1/(1-s)$$
$$s = (\ln(\tau_{SR2})- \ln(\tau_{SR1}))/(\ln(B_{SR2})-\ln(B_{SR1}))$$

where B is the total atmospheric CH_4 burden. F describes the response of the atmospheric CH_4 abundance to a change in CH_4 emissions. In the case of a small perturbation, F is approximately the ratio of the relative change in CH_4 concentrations to an imposed emission change; for example, the model ensemble mean F of 1.33 (Table 2) implies that a 1% increase in CH_4 emissions would ultimately yield a 1.33% increase in CH_4 concentrations. The multi-model mean F is at the low end of the 1.33 to 1.45 range reported (and within 10% of the recommended F of 1.4) by *Prather et al*. [2001].

Following *Naik et al*. [2005] and *West et al*. [2007], we estimate the long-term impact on O_3 by scaling the change in surface O_3 in the CH_4 perturbation simulation (SR2-SR1) by the ratio of the estimated changes in CH_4 from SR1 to SRNxx versus SR2:

$$\Delta O_3 (SRNxx – SR1) = [\Delta CH_4 (SRNxx – SR1)/ \Delta CH_4 (SR2\text{-}SR1)] * \Delta O_3 (SR2\text{-}SR1)$$

For each model, we add this ΔO_3 to the short-term O_3 response diagnosed directly from SRNxx-SR1.

Including the long-term feedback through CH_4 has little impact on the model ensemble mean domestic response (solid vs. dotted lines in Figure 4). In contrast, a larger percentage change occurs for the O_3 response to foreign emissions since the O_3 response to changes in CH_4 (SR2-SR1) is relatively uniform globally. Figure 5 shows that the long-term contribution diminishes the estimated O_3 response to foreign NO_x emissions by ~15-20% during the month of maximum foreign contribution (April for NA, EU, and EA and November for SA) since decreasing NO_x lowers OH, causing the CH_4 abundance to rise, thereby enhancing the CH_4 contribution to surface O_3. During the same months, the short-term O_3 responses to the 20% reductions in foreign CO and NMVOC emissions are augmented by 30-40% and ~10%, respectively, since decreasing

CO or NMVOC increases OH. These results are qualitatively consistent with those of *West et al*. [2007]. Over NA and EU during summer, the opposing influences of the long-term feedback from NO_x and CO result in the total impact of the CO emission reductions exceeding that from NO_x (or NMVOC). As there is little seasonality in the O_3 response to CH_4 (SR2-SR1), the seasonal cycle of the total O_3 response is mainly driven by the short-term response to the changes in NO_x, CO, and NMVOC emissions (Figure 6).

In the case of "ALL", the long-term feedback is minimal (always less than 3% for all months and regions in Figure 5). The balancing effect of simultaneous changes of NO_x, CO, and NMVOC has been noted before in the context of the remarkable stability of OH concentrations from the pre-industrial to the present-day atmosphere [*Wang and Jacob*, 1998; *Lelieveld et al*., 2002b]. Since anthropogenic sources of NO_x, CO, and NMVOC differ, however, equivalent percentage reductions would not necessarily be applied to all precursors together, in which case the long-term effect should be considered.

5.*3 Inferring the O_3 response to regional reductions in anthropogenic CH_4 emissions*

Since the global CH_4 abundance was decreased uniformly by 20%, the results from this simulation are not directly comparable with those from the 20% regional reductions of the other O_3 precursors. In this section we attempt such a comparison by approximating the surface O_3 response that would result from 20% reductions of CH_4 anthropogenic emissions in the source regions.

We first use ensemble mean results to estimate the anthropogenic CH_4 emission decrease that would produce the 20% reduction in global concentrations applied in the SR2 simulation. Applying the model ensemble mean feedback factor (F) of 1.33 from Table 2 (Section 5.1) to account for the feedback of CH_4 on its own lifetime, we derive that the 20% decrease in CH_4 abundance corresponds to a 15% decrease in total global CH_4 emissions. Assuming that anthropogenic CH_4 emissions are 60% of the total CH_4 emissions [*Denman et al.*, 2007], this 15% decrease in total global CH_4 emissions corresponds to a 25% decrease in global anthropogenic CH_4 emissions. We then use the EDGAR 3.2 FT2000 anthropogenic CH_4 emission inventory [*Olivier et al.*, 2005] to estimate that NA, EU, SA, and EA each contribute 16.6%, 16.0%, 17.3%, and 19.0%, respectively, to total global anthropogenic emissions (298 Tg CH_4 yr^{-1} in 2000); together the anthropogenic CH_4 emissions from these four regions contribute 68.9 % to global total anthropogenic emissions.

The fraction of the total O_3 response diagnosed from SR2-SR1 that would be produced by 20% decreases in regional anthropogenic CH_4 emissions can then be estimated (*i.e*., for NA: (0.2*16.6% of global anthropogenic emissions)/(25% decrease in global anthropogenic emissions as implied by the concentration change in SR2), assuming that the O_3 response scales linearly with changes in anthropogenic CH_4 emissions and does not depend on the location of the CH_4 emission reductions [*Fiore et al.*, 2008]. We obtain values of 13.3%, 12.8%, 13.8%, and 15.2% of the O_3 decrease in the SR2 simulation for NA, EU, SA, and EA, respectively. We scale the surface O_3 response to CH_4 over the receptor regions (SR2-SR1 in Table 2) by these values to

estimate an annual mean surface O_3 decrease for each SR pair (Table A6) that ranges from 0.14 ppb (NA and EU on EA) to 0.20 ppb (EA on SA).

The results are shown in Figure 3 for comparison with the O_3 decreases achieved with reductions in the regional emissions of the traditional O_3 precursors (NO_x, NMVOC, and CO). While the combined domestic emission reductions of the traditional O_3 precursors ("ALL" in Figure 3) are most effective at reducing surface O_3 over all regions, the additional inclusion of domestic CH_4 emission reductions would yield another 14-20% decrease in annual mean surface O_3. Comparison of the "3 foreign" bars (black) in "ALL" versus "CH_4" in Figure 3 implies that the inclusion of anthropogenic CH_4 emissions in a multi-species control strategy to reduce background surface O_3 in the Northern Hemisphere would nearly double the surface O_3 decrease attained by controlling the traditional O_3 precursors alone. The larger O_3 response over foreign regions to anthropogenic CH_4 versus "ALL" O_3 precursor emissions from SA reflects the comparable amounts of anthropogenic CH_4 emissions from the four regions, whereas SA emits only half as much NO_x as the other regions. For 6 SR pairs, regional anthropogenic CH_4 emission reductions are estimated to yield equivalent (within ±25%) responses in surface O_3 over foreign continents as the coincident reductions in traditional O_3 precursors (ALL). For NA on EU, the influence from regional CH_4 emission reductions is roughly half that of the traditional O_3 precursors, and for three SR pairs (NA on EA and EU on EA and SA), the CH_4 response is 30-40% less than that to "ALL". The additional hemispheric-wide O_3 decrease from reductions in anthropogenic CH_4 emissions would occur on the time scale of the CH_4 lifetime, approximately a decade, whereas the response to the traditional O_3 precursors will occur in weeks to months.

6. Response of air quality as measured by threshold statistics

In many nations, compliance with air quality standards is assessed with respect to a threshold concentration. *Ellingsen et al*. [2008] have shown that threshold statistics based on 35, 60, and 80 ppb O_3, simulated by global CTMs, responded similarly to changes in precursor emissions across 14 world regions ($r^2 > 0.55$). Here we focus on incidences of daily maximum 8-hour average O_3 concentrations above 60 ppb (DAYS>60), a statistic used in Europe to protect human health, with a target value of 25 days or fewer per year [*e.g.,* as discussed further by *Ellingsen et al.*, 2008]. We previously showed that the model ensemble mean closely matches the monthly average O_3 observations over EU (Figure 2), and so we focus on the EMEP sites (Figure 1) to compare observed and simulated DAYS>60; results are shown in Figure 12. Over the Mediterranean and low-altitude (below 1 km) Central European regions several models (and the model ensemble mean and median) are within the observed range of DAYS>60. Consistent with the results in Figure 2, Figure 12 shows that the models tend to underestimate DAYS>60 at the high-altitude central Europe with only 2 models simulating values within the observed range. We recommend further work to determine why some models capture this statistic better than others, including the potential role of differences in mixed layer depths and deposition.

We next use the model ensemble mean values to explore the relevance of the monthly mean results in Figure 6 for threshold metrics. Figure 13 shows the model ensemble domain average monthly mean DAYS>60 in the base simulation (right axis) for

each region, and the change in these values when all O_3 precursors are decreased by 20% in the foreign source regions (left axis). In the base simulation, DAYS>60 peaks in summer over NA and EU, in spring over EA, and in winter to early spring over SA (Figure 13). The response of DAYS>60 to the foreign emission reductions is strongest in spring (decreases of 5-10% of the base case for some SR pairs), with the low incidence of DAYS>60 in late fall preventing the secondary maximum shown in Figure 6 from emerging. The rankings of the O_3 responses to foreign emission decreases in spring are consistent with those in Section 4, suggesting that the mean response qualitatively describes the response of the threshold statistics relevant for gauging attainment of air quality standards. We emphasize that the values in Figure 13 are averaged over large spatial areas and mask a large variability within the regions; for example, the model ensemble mean DAYS>60 over Europe decrease by 2-4 days over much of Southern Europe and Northern Africa, and by more than 5 days over the Middle East when ALL emissions are decreased by 20% in NA (not shown). A forthcoming manuscript [*Reidmiller et al.*] will expand this analysis further to examine variability within the United States.

The total annual (spatial average) decrease in DAYS>60 attained with foreign emission reductions is <10-20% of the decrease from equivalent percentage reductions applied domestically (not shown), smaller than for the annual mean changes in Section 4. This result implies that DAYS>60 (and thus higher O_3 values) are more sensitive to domestic emissions, with the highest O_3 levels occurring during meteorological conditions favoring regional production from domestic emissions.

7. Consistency of O_3 trends derived from measurements versus models

Observational evidence indicates that background surface O_3 at northern mid-latitudes has been increasing by 0.1-0.5 ppb yr^{-1} in recent decades [*e.g. Vingarzan*, 2004; *Carslaw*, 2005; *Jonson et al*., 2005; *Derwent et al.*, 2007; *Jaffe and Ray* 2007] although estimated trends vary and may even be leveling off [*Oltmans et al*., 2006; *Derwent et al*., 2007]. The model ensemble mean O_3 SR relationships diagnosed here can be combined with reported anthropogenic emission trends to estimate emission-driven changes in Northern Hemispheric background surface O_3 for comparison with trends derived from observations. While wintertime O_3 increases over Europe have been mainly attributed to decreases in EU NO_x emissions [*Jonson et al*., 2005; *Ordóñez et al*., 2005], we consider here the increase observed throughout the year, and examine the consistency of the multi-model ensemble mean SR relationships with the hypothesis that a 0.1-0.5 ppb yr^{-1} increase in Northern Hemispheric background O_3 may be driven by precursor emissions associated with rapid industrialization in Asia [*e.g. Jaffe et al*. 2003c; *Parrish et al*., 2004]. Recent satellite retrievals of NO_2 columns suggest that NO_x emissions from China have increased by ~40%, with the growth rate accelerating from 4 to 12% per year, from 1996 to 2002 [*Richter et al*., 2005]. During this same period, little change is found over the United States, while the satellite NO_2 columns suggest a decrease of 30% over western Europe [*Richter et al*., 2005].

The annual mean surface O_3 response to EA anthropogenic NO_x emissions from Table A4 (~0.1 ppb over the foreign regions) implies that a +10% yr^{-1} increase in EA NO_x emissions would yield an O_3 response of ~0.05 ppb yr^{-1}, below the observed range.

If we instead assume that all Asian (i.e., EA+SA) emissions are increasing together with NO_x, the 20% emission reductions in Asia produce a ~0.2 (over EU) to 0.3 ppb (over NA) O_3 decrease (ALL in Table A4). In this case, the 10% yr^{-1} increase in Asian O_3 precursor emissions scales to an O_3 response that falls within the lower end of the surface O_3 increase derived from observations: a 0.1 and 0.15 ppb yr^{-1} increase in O_3 averaged over the EU and NA receptor regions, respectively. Considering the decreases in European NO_x emissions (~5% yr^{-1}), we estimate a 0.02 and 0.03 ppb yr^{-1} annual mean decrease in surface O_3 over NA and EA, respectively, or 0.04 and 0.05 ppb yr^{-1} if EU CO and NMVOC emissions follow the EU NO_x trend. We conclude that the Asian NO_x emission changes estimated by *Richter et al.* [2005], if accompanied by increases in the other O_3 precursors over Asia, are consistent with annual mean O_3 trends of ~0.1 ppb yr^{-1}, but insufficient to produce a 0.5 ppb yr^{-1} trend. In a similar manner, we estimate that the 60% increase in South Asian NO_x emissions projected by the Current Legislation (CLE) emission scenario between 2005 and 2030 [*Dentener et al*., 2005; *Cofala et al*., 2005] resulting from growth in the power and transportation sectors would increase surface O_3 over NA, EU, and EA by less than 0.3 ppb.

The ability of current models to attribute fully the observed increases in background O_3 at northern mid-latitudes deserves further attention, particularly given a growing demand for future projections of climate- and emission-driven changes in surface O_3. Our assumptions that the NO_x trends reported by *Richter et al.* [2005] apply to emissions of other O_3 precursors as well as the SA region would cause us to overestimate the resulting surface O_3 trend. On the other hand, the responses diagnosed here are spatially averaged over large continental regions and should be biased low compared to trends derived from measurements at remote sites that are often situated on the western coasts of North America and Europe, or on mountain summits where they sample free tropospheric air. We expect the intercontinental signal at such sites to be larger than in surface air over regions where O_3 is subjected to higher depositional and chemical loss rates. In addition to rising anthropogenic emissions, regional changes in climate, biogenic emissions and wildfires may contribute to the observed O_3 trends [*e.g.*, *Jaffe and Ray*, 2007*; Jaffe et al*., 2008].

8. Conclusions

Under the umbrella of the Task Force on Hemispheric Transport of Air Pollution (TF HTAP; www.htap.org), we have used an ensemble modeling approach to estimate source-receptor (SR) relationships for surface O_3 and its precursor emissions from four continental regions at northern mid-latitudes (Figure 1). Specifically, 21 global and hemispheric chemical transport models used meteorology for 2001 to simulate the impact of 20% decreases in "conventional" O_3 precursor emissions (NO_x, NMVOC, and CO individually and combined) from East Asia (EA), Europe (EU), North America (NA) and South Asia (SA) on surface O_3 in the same four regions. Our results are intended to provide a first comprehensive assessment of annual and seasonal mean intercontinental SR relationships, to gauge uncertainty in these estimates, and to serve as a benchmark for future work. The consistent approach applied in our study narrows the wide range of SR relationships reported in the literature (Figure 11). We identified a systematic model

826 overestimate of surface O_3 concentrations compared to observations over the eastern
827 United States and Japan, and a strong sensitivity to uncertainties in anthropogenic
828 NMVOC emissions, particularly over EU. Future work should determine the extent to
829 which our results apply to other meteorological years, and explore sub-continental
830 variability in SR relationships driven by differences in the availability of O_3 precursors
831 and in meteorological conditions [*e.g.*, *Holloway et al*., 2007].
832 In addition to the precursors that are traditionally regulated to abate O_3 pollution
833 (*i.e.*, NO_x, NMVOC, and CO), we examined the contribution of CH_4 to hemispheric-wide
834 surface O_3 levels, both directly by changing global CH_4 abundances, and indirectly
835 through the influence that the traditional O_3 precursors have on OH concentrations, and
836 thereby the CH_4 abundance [*e.g. Prather*, 1996]. Due to competing effects of CO and
837 NMVOC versus NO_x on OH, neglecting the long-term feedback when equivalent
838 percentages of CO, NMVOC and NO_x are reduced together introduces errors of at most a
839 few percent. Given the different anthropogenic sources of NO_x, CO, and NMVOC,
840 application of equivalent percentage reductions to all three precursors may not be
841 pragmatic, in which case the long-term effect may not be trivial. We further show that
842 the responses to single-component versus multi-component emission reductions are
843 approximately equivalent for the O_3 responses to changes in domestic and foreign
844 emissions, although the multi-component response is less-than-linear in coupled aerosol
845 simulations in which aerosols and their precursors were also decreased. We focus the
846 remainder of our conclusions on the simulations with the combined reductions of NO_x,
847 CO, and NMVOC.
848 For all source regions, the model ensemble mean intercontinental influence is
849 largest during spring and in late fall (Figure 6), consistent with prior studies (Figure 11
850 and Section 1). The most robust rankings across the models are that surface O_3 levels
851 over EU are influenced most strongly by emission reductions in NA, followed by EA,
852 and that SA contributes least to the three foreign regions. We find more uncertainty in
853 the relative importance of EA versus EU on NA, and of the three source regions over
854 both Asian regions. Our analysis of the impact of decreases in anthropogenic emissions
855 on the incidence of daily maximum 8-hour average O_3 concentrations above 60 ppb
856 suggests that the annual and seasonal mean responses are qualitatively relevant for
857 assessing air quality changes as measured by a threshold statistic used to gauge
858 compliance with air quality standards.
859 We define an "import sensitivity" as the ratio of the sum of the change in surface
860 O_3 resulting from perturbations to precursor emissions in the three foreign source regions
861 to the surface O_3 change resulting from the same perturbations to domestic emissions.
862 The model ensemble mean import sensitivity ranges from 0.5 (SA in November) to
863 1.1(EA in March) during the month with the largest surface O_3 response to the combined
864 foreign emission reductions. Regional O_3 production reaches a maximum in summer
865 over EA, EU, and NA (Figure 5), with model ensemble mean import sensitivities for July
866 of 0.2 (NA and EA) to 0.3 (EU). We assign a high degree of uncertainty, however, to the
867 NA and EA results given the model bias compared to surface observations during the
868 summer and early fall (Figure 2).
869 The responses to emission reductions of the traditional O_3 precursors from a
870 single foreign source region are often 10% (maximum of ~50%) of the responses to
871 domestic emission reductions. This foreign influence is not trivial if one considers that

application of the cheapest air pollution control measures in industrialized nations renders additional controls disproportionately expensive [*Keating et al*., 2004] and that our results are large spatial averages that do not convey the larger foreign influence occurring in some sub-continental regions (*e.g.* the west coasts of NA and EU). From the multi-model ensemble mean response to a 20% decrease in global CH_4 abundances, we infer that the O_3 decrease over foreign regions produced by regional reductions in anthropogenic CH_4 emissions is roughly equivalent to the O_3 decrease from the same percentage reduction of NO_x, NMVOC, and CO together. The inclusion of anthropogenic CH_4 emissions as a strategy to decrease hemispheric O_3 may be economically attractive given the availability of low-cost control measures [*West and Fiore*, 2005]. From a perspective of jointly addressing climate and air quality, a multi-species approach to reducing O_3 pollution is preferable to reducing NO_x alone [e.g., *Wild et al.*, 2001; *Fuglestvedt et al.*, 1999, *Naik et al.* 2005; *Shindell et al.*, 2005]. Our results provide a baseline for future assessments of intercontinental SR relationships. This baseline is a key step towards informing air pollution managers confronted by the possibility that future increases in global emissions could offset air quality improvements attained via regulations on domestic emissions.

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FIGURE CAPTIONS

Figure 1. The HTAP source-receptor regions: NA (15-55°N; 60-125°W), EU (25-65°N; 10°W-50°E) EA (15-50°N; 95-160°E), and SA (5-35°N; 50-95°E). Sites marked with the same symbols are used to produce the sub-regional averages in Figure 2, from the Co-operative programme for monitoring and evaluation of the long-range transmissions (EMEP) in the Mediterannean (red diamonds; panel a in Figure 2) and Central Europe (green open triangles for sites below 1 km and blue crosses for sites > 1 km; panels b and c in Figure 2, respectively); from the U.S. Clean Air Status and Trends Network (CASTNet) in the Northeast (red circles; panel d in Figure 2) Southwest (green triangles; e), Southeast (dark blue inverted triangles; f), Great Lakes (pink diamonds; g), Mountainous West (cyan squares; h), and from the Acid Deposition Monitoring Network in East Asia (EANET) in Japan (red asterisks; panel i in Figure 2).

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Figure 5. Same as Figure 4, but for the sum of the O_3 responses to 20% decreases in anthropogenic emissions in the three foreign source regions.

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Figure 9. Change in surface O_3 over the EU (left) and NA (right) as a function of various size perturbations to EU anthropogenic NO_x emissions by season (colors), as simulated with the FRSGC/UCI model (solid lines with circles) and as estimated by scaling linearly from the response in the simulation where NO_x emissions were decreased by 20% (SR3EU; dotted lines).

Figure 10. Decrease in annual mean NA surface O_3 (ppb) resulting from 20% reductions in EU anthropogenic NMVOC emissions (SR1-SR4EU) plotted against the EU anthropogenic NMVOC emissions (Tg C a^{-1}). The points represent the results from individual models (EU NMVOC emissions are given in Table A3). The coefficient of determination (r^2) is 0.50.

Figure 11. Annual and seasonal mean contribution to total surface O_3 from foreign source regions as estimated from the individual model results in this study (colored by source region: green for EU; blue for EA, grey for EA+EU; red for NA) and from studies in the published literature (thin vertical bars for range across studies and regions; squares where one value is reported; note that regional definitions, methods for source attribution, and reported metrics (e.g. 24-hour vs. afternoon vs. daytime mean) vary across studies) [*Derwent et al*., 1998; *Berntsen et al*., 1999; *Wild and Akimoto*, 2001; *Derwent et al*.,

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Figure 12: Observed (black circles) and simulated (grey diamonds) annual number of days when daily maximum 8-hour average O_3 concentrations exceed 60 ppb at the EMEP stations, averaged over the regions in Figure 2: Mediterranean (Medit.), Central European sites below 1 km altitude (C EU < 1km) and above 1 km altitude (C EU > 1km). The black vertical bars depict the standard deviation of the observed values across the stations within the region. The model ensemble mean (red circles) and median (green circles) values from the 18 models that contributed hourly surface O_3 results for SR1 are also shown.

Figure 13. As in Figure 6 but for the model ensemble mean change in the number of days per month when daily maximum 8-hour average O_3 concentrations exceed 60 ppb, spatially averaged within each receptor region (panels). The change is estimated by first calculating the area-weighted spatial average value for DAYS>60 in each model simulation for a given region (the right axis shows DAYS>60 in the base case, SR1), and then taking the multi-model average of the differences in the spatial average DAYS>60 values (SR6-SR1) from the 12 models that provided hourly surface O_3 results for SR6.

Table 1. Model ensemble annual mean (median) ± one standard deviation in total and anthropogenic emissions of NO_x (Tg N a^{-1}), NMVOC (Tg C a^{-1}), and CO (Tg a^{-1}), globally and for the regions in Figure 1. Emissions in individual models are provided in Tables A1 and A2.

Total Emis.	GLOBAL	NA	EU	EA	SA
NO_x	46.5(46.2)±5.7	8.5(8.7)±0.8	8.4(8.4)±1.1	7.1(6.9)±1.4	3.3(3.3)±0.5
NMVOC	630(623)±221	62(57)±24	37(34)±13	48(47)±14	33(34)±8.8
CO	1060(1090)±135	130(130)±20	90(81)±25	150(150) ±29	97(96)±23
Anthr. Emis.					
NO_x	32.5(29.4)±6.0	7.4(7.3)±0.4	7.3(7.5)±0.6	6.0(5.5)±1.4	2.4(2.2)±0.4
NMVOC	96.8(92.3)±41.8	16(16)±7.0	19(20)±11	16(17)±6.5	10(10)±3.9
CO	661(563)±214	101(103)±19	80(70)±23	133(123) ±35	80(79)±18

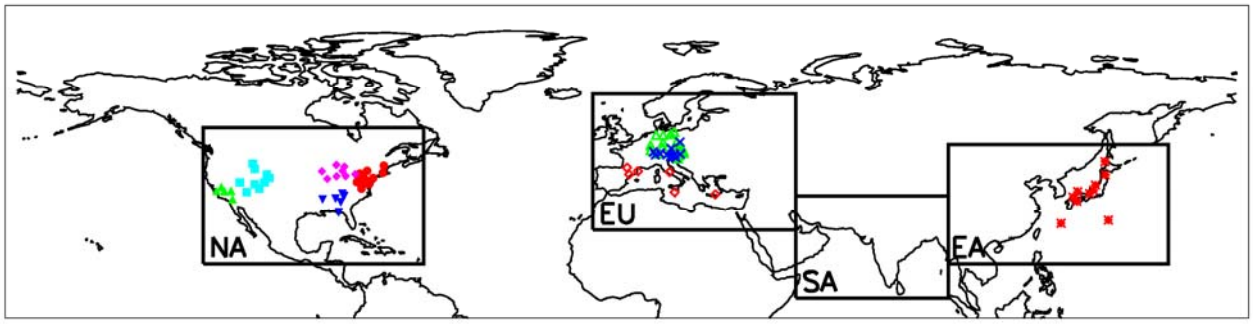
Table 2. Methane lifetime (τ) and "feedback factor" (F) in the individual models

MODEL	τ_{OH}[a]	τ_{tot}[b]	F[c]
CAMCHEM-3311m13	11.86	10.11	1.30
FRSGCUCI-v01	8.70	7.72	1.43
GISS-PUCCINI_modelE	10.88	9.39	1.36
GMI-v02f	10.38	9.02	1.31
LMDz3-INCA1	10.02	8.74	1.31
LLNL_IMPACT-T5a	6.19	5.68	1.40
MOZARTGFDL-v2	10.44	9.06	1.31
MOZECH-v16	11.20	9.63	1.29
STOCHEM-HadGEM	11.72	10.01	1.28
TM5-JRC-cy2-ipcc-v1	9.02	7.97	1.43
UM-CAM-v01	12.50	10.57	1.25
Model Ensemble Mean	**10.26**	**8.90**	**1.33**
Standard Deviation	**1.78**	**1.38**	**0.06**

[a] The methane lifetime against loss by tropospheric OH (years), defined as the total atmospheric burden divided by the tropospheric CH_4 loss rates, with the troposphere defined using the 150 ppb O_3 chemical tropopause.

[b] τ = total atmospheric CH_4 lifetime (years) determined from τ_{OH} and assuming CH_4 losses to soils and the stratosphere with lifetimes of 160 and 120 years [*Prather et al.*, 2001], respectively.

[c] The feedback factor is the ratio of the atmospheric response (or perturbation) time to the global atmospheric lifetime and is given by 1/(1-s) where s is determined from the SR2 and SR1 simulations, and defined as $\delta \ln(\tau) / \delta \ln[CH_4]$ [*Prather et al.*, 2001], where $[CH_4]$ = 1760 ppb in SR1 and 1408 ppb in SR2.



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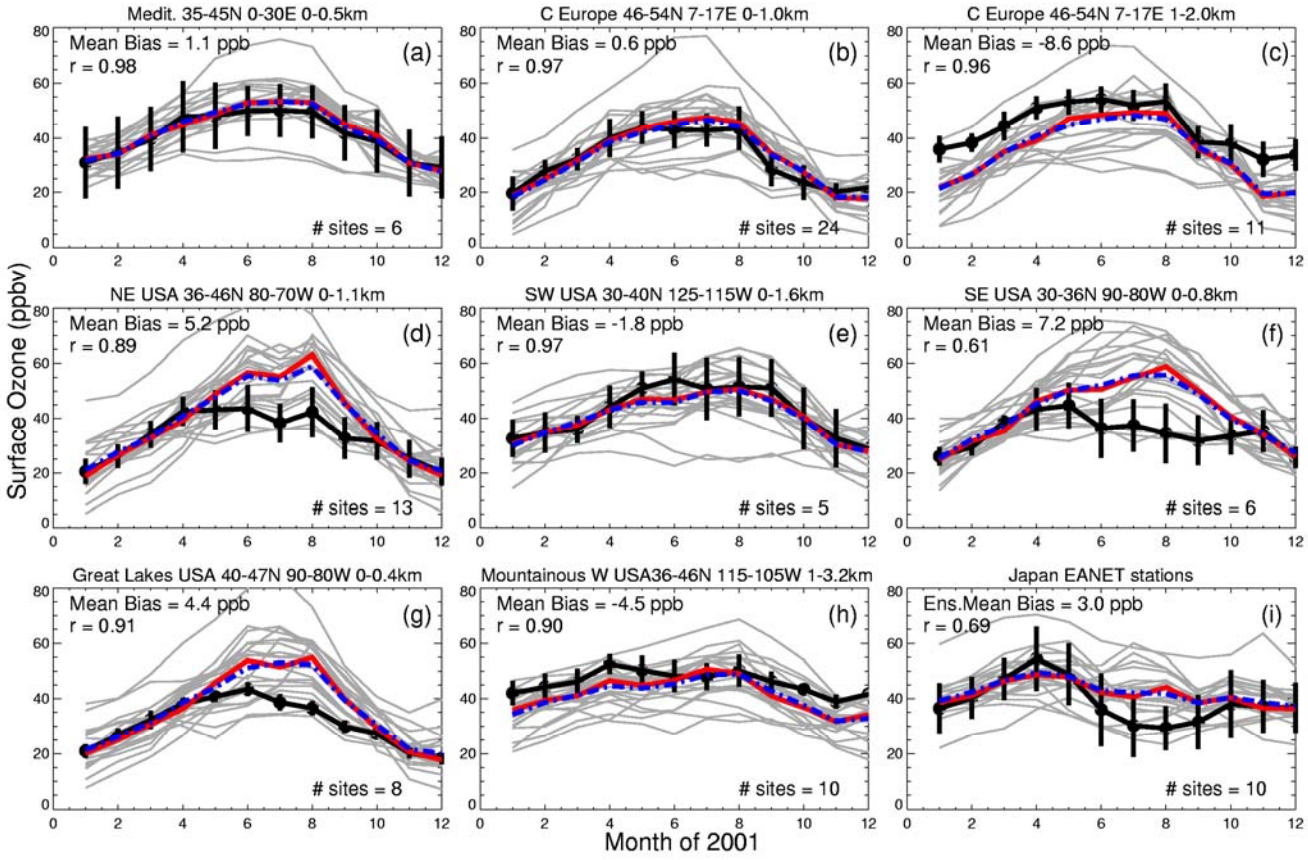

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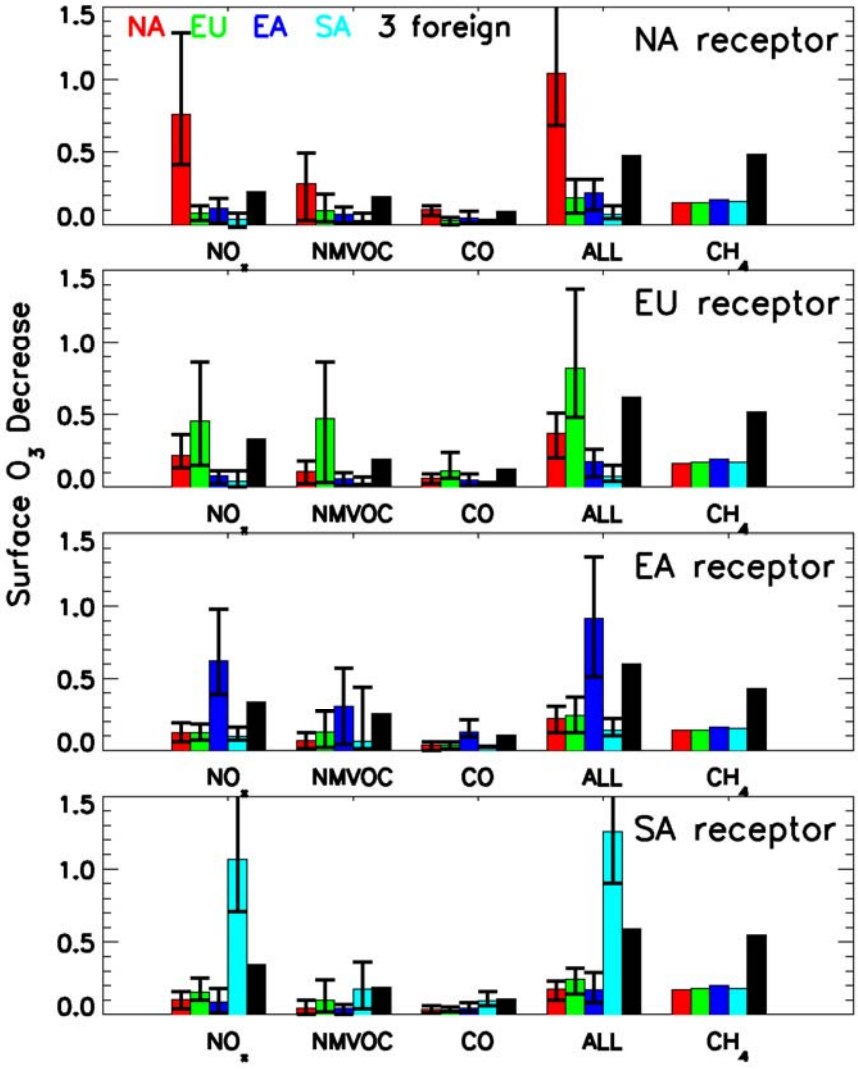

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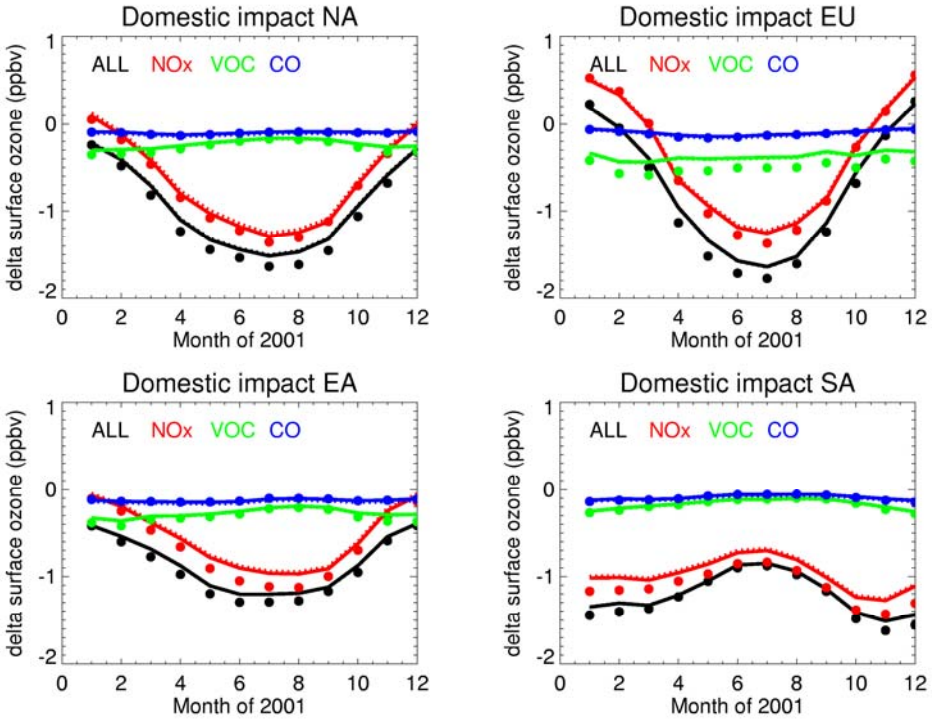

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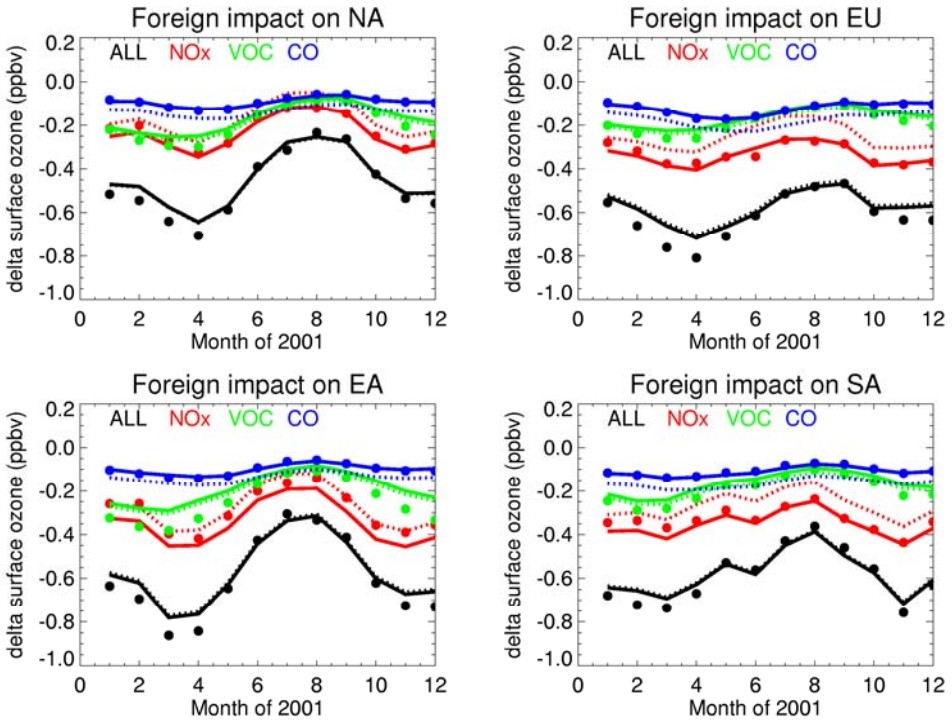

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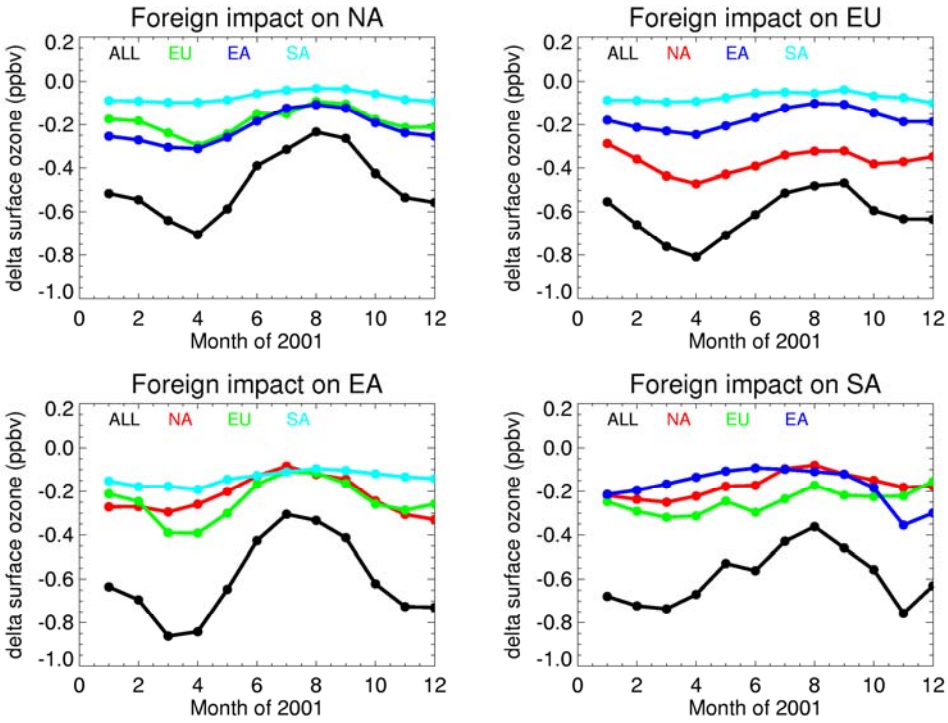

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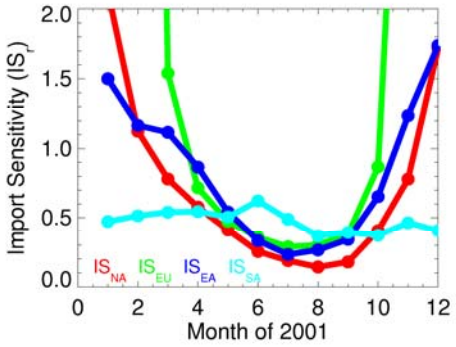

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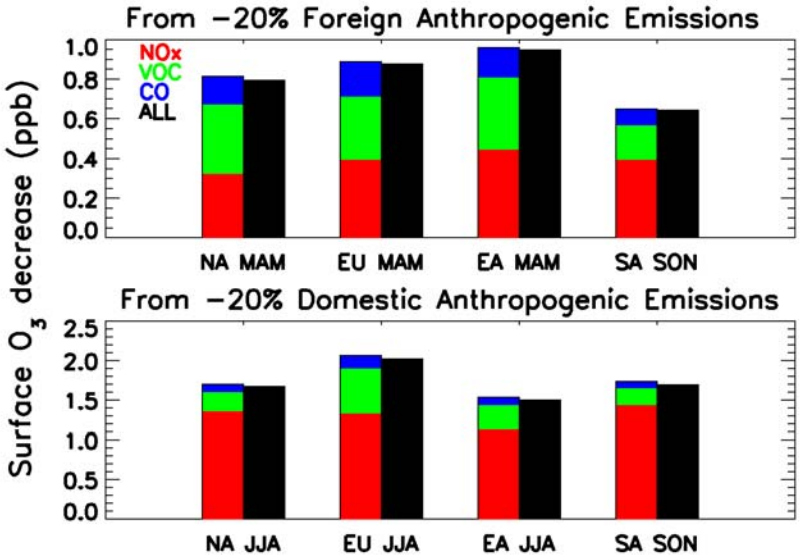

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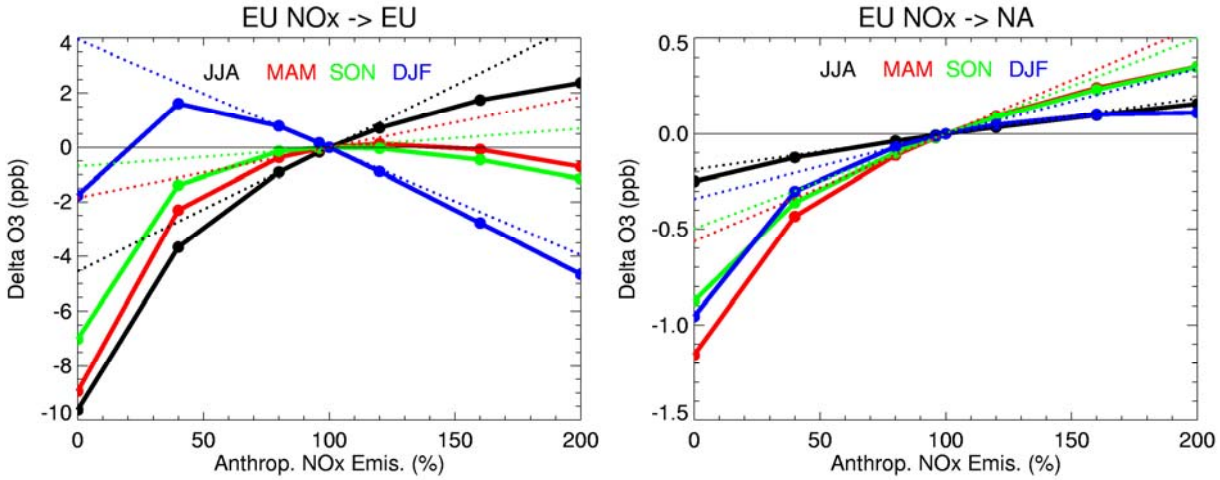

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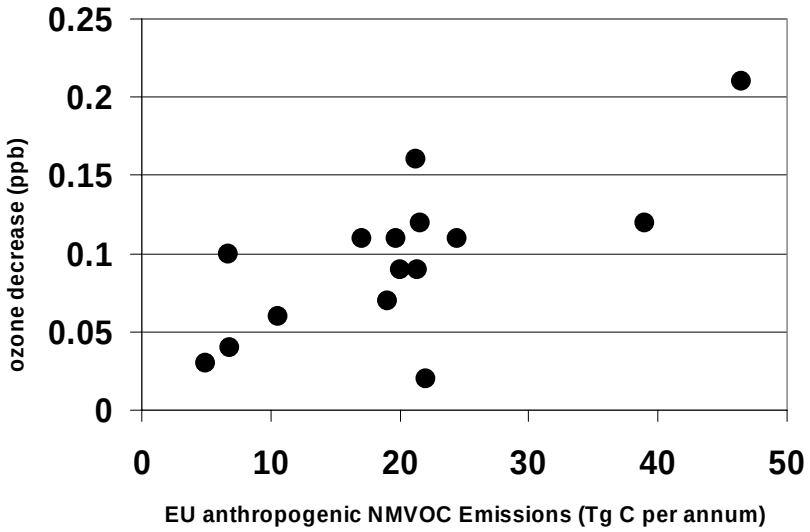

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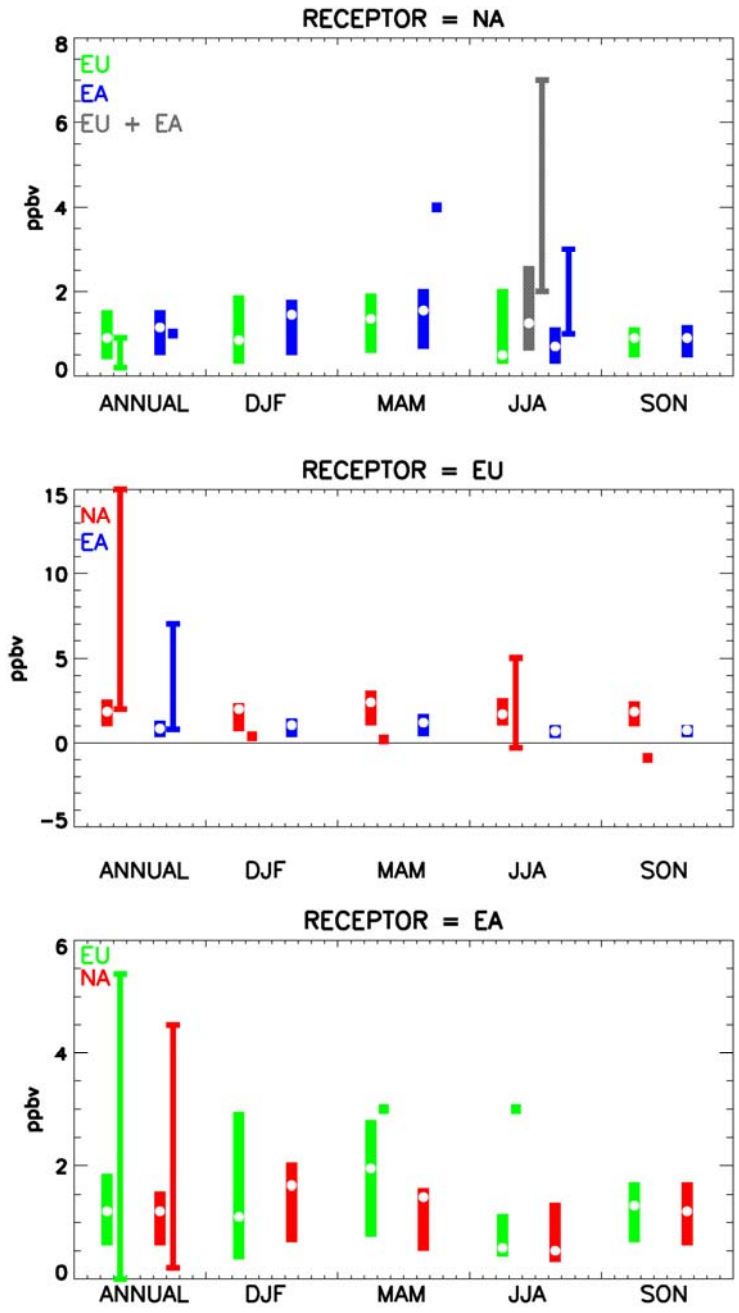

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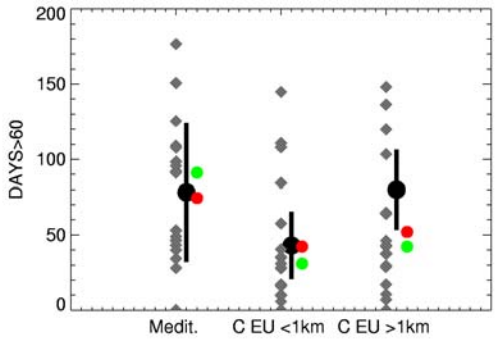

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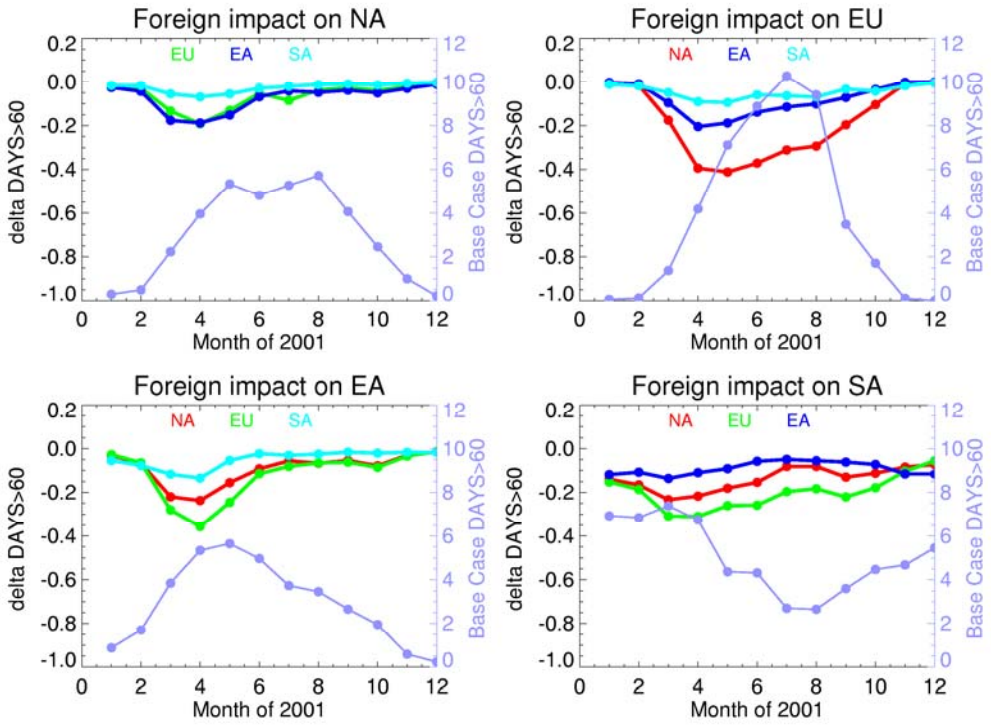

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