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Effect of regional precursor emission controls on long-range ozone transport – Part 1: Short-term changes in ozone air quality

J. J. West[1], **V. Naik**[2,*], **L. W. Horowitz**[3], **and A. M. Fiore**[3]

[1]University of North Carolina, Chapel Hill, NC, USA
[2]Princeton University, Princeton, NJ, USA
[3]NOAA Geophysical Fluid Dynamics Laboratory, Princeton, NJ, USA
[*]now at: ATMOS Research and Consulting, Lubbock, TX, USA

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Abstract. Observations and models demonstrate that ozone and its precursors can be transported between continents and across oceans. We model the influences of 10% reductions in anthropogenic nitrogen oxide (NO_x) emissions from each of nine world regions on surface ozone air quality in that region and all other regions. In doing so, we quantify the relative importance of long-range transport between all source-receptor pairs, for direct short-term ozone changes. We find that for population-weighted concentrations during the three-month "ozone-season", the strongest inter-regional influences are from Europe to the Former Soviet Union, East Asia to Southeast Asia, and Europe to Africa. The largest influences per unit of NO_x reduced, however, are seen for source regions in the tropics and Southern Hemisphere, which we attribute mainly to greater sensitivity to changes in NO_x in the lower troposphere, and secondarily to increased vertical convection to the free troposphere in tropical regions, allowing pollutants to be transported further. Results show, for example, that NO_x reductions in North America are ∼20% as effective per unit NO_x in reducing ozone in Europe during summer, as NO_x reductions from Europe itself. Reducing anthropogenic emissions of non-methane volatile organic compounds (NMVOCs) and carbon monoxide (CO) by 10% in selected regions, can have as large an impact on long-range ozone transport as NO_x reductions, depending on the source region. We find that for many source-receptor pairs, the season of greatest long-range influence does not coincide with the season when ozone is highest in the receptor region. Reducing NO_x emissions in most source regions causes a larger decrease in export of ozone from the source region than in ozone production outside of the source region.

Correspondence to: J. J. West
(jasonwest@unc.edu)

1 Motivation

Tropospheric ozone is a pervasive air pollutant that affects human health, crop productivity, and natural ecosystems. Ozone is also a major greenhouse gas, and changes in emissions of ozone precursors – nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs), carbon monoxide (CO), and methane (CH_4) – affect the global climate through a linked chemical system (Forster et al., 2007).

Ozone has traditionally been addressed as a local air pollutant, through reductions in emissions of NMVOCs and NO_x. Over the past two decades, ozone has been increasingly recognized as a regional pollutant, and NO_x emission reductions have received greater emphasis (National Research Council, 1991). More recently, it has been shown that anthropogenic emissions of ozone precursors affect ozone concentrations over larger spatial scales, including between continents, as demonstrated through direct surface observations, satellite measurements, and modeling studies (see Akimoto, 2003; Holloway et al., 2003; Stohl, 2004; Task Force on Hemispheric Transport of Air Pollution, 2007; and references therein). Observations and modeling studies have likewise shown that global background concentrations of ozone have increased since preindustrial times, including at the surface (Vingarzan, 2004). Growth in this global background has been attributed to anthropogenic emissions of ozone precursors, mainly NO_x and CH_4 (Wang and Jacob, 1998; Lelieveld and Dentener, 2000).

The inter-continental transport of ozone results from both the formation of ozone over source regions, with subsequent transport, and the transport of ozone precursors. Because the lifetime of ozone in the troposphere (days to months, with a tropospheric mean of ∼22 days, Stevenson et al., 2006) is generally much longer than typical transport times between continents separated by oceans (5–10 days), the inter-continental transport of ozone may be substantial. The

lifetime of ozone is also generally much longer than that of NO_x (roughly 1–2 days), which would suggest that the transport of ozone dominates over the transport of NO_x. However, ozone formation in remote regions of the troposphere is commonly limited by NO_x, and therefore the fraction of NO_x emissions that is transported out of polluted continental regions can be relatively more effective at producing ozone. Further, a fraction of NO_x from polluted source regions is converted to reservoir species, such as the peroxyacyl nitrates (PANs), which persist longer in the free troposphere before subsiding and decomposing to give NO_x, thereby producing ozone further from the sources (Moxim et al., 1996; Wang et al., 1998; Horowitz and Jacob, 1999).

Previous modeling studies of the inter-continental transport of ozone and its precursors have mainly considered transport between two geographical regions, such as the influence of Asian emissions on air quality in North America (Jacob et al., 1999; Berntsen et al., 1999; Yienger et al., 2000; Hudman et al., 2004; Zhang et al., 2008), North America and Europe (Li et al., 2002), and Europe and Asia (Wild et al., 2004). Other studies have addressed the pathways for the export of pollution from or import to populated regions of interest (Stohl, 2004), including the export from the USA (Liang et al., 1998), Europe (Duncan and Bey, 2004; Duncan et al., 2008), and Asia (Newell and Evans, 2000; Phadnis and Carmichael, 2000; Mauzerall et al., 2000; Heald et al., 2003; Liu et al., 2003; Kunhikrishnan et al., 2004; Liu et al., 2005), and the import of pollution to Europe (Stohl and Trickl, 1999; Trickl et al., 2003; Stohl et al., 2003; Derwent et al., 2004, 2008), the US (Fiore et al., 2002, 2003; Lin et al., 2008), and Asia (Kunhikrishnan and Lawrence, 2004; Kunhikrishnan et al., 2006; Holloway et al., 2008).

Because these studies have used different models under different conditions with different experimental designs, it is difficult to compare the results to quantify the relative strength of ozone transport between different source and receptor regions (Task Force on Hemispheric Transport of Air Pollution, 2007). Few studies have considered multiple regions simultaneously in a common modeling framework, such that these influences can be compared directly. Fuglestvedt et al. (1999), Berntsen et al. (2005), and Naik et al. (2005) simulated changes in ozone precursor emissions from several world regions to assess their effects on tropospheric ozone and radiative climate forcing, but do not present results for surface air quality. Wild and Akimoto (2001) simulated the transport of ozone between three regions (USA, Europe, and East Asia) due to 10% increases in emissions of NO_x, CO, and non-methane hydrocarbons from fossil fuel sources. Sudo and Akimoto (2007) used tagged tracer simulations to track the global distributions of ozone based on its region of formation, for many source regions globally. Hess and Lamarque (2007) similarly used tagged emissions to study the transport of ozone in the Northern Hemisphere (NH) and its variability with the Arctic Oscillation. Recently, a large model intercomparison study was conducted, considering the transport of ozone among four source regions by simulating emission reductions (Task Force on Hemispheric Transport of Air Pollution, 2007; Fiore et al., 2009).

Here we consider nine world regions, and use a global model of atmospheric chemistry and transport to identify the strongest linkages, for surface ozone air quality, among all combinations of source and receptor regions. We estimate the influence of a 10% reduction in anthropogenic NO_x emissions from each of these nine regions individually, considering the effects on surface ozone air quality within that region and in all other regions. Simultaneous 10% reductions of anthropogenic NO_x, NMVOC, and CO emissions are also simulated for three source regions. We present results for several surface ozone air quality metrics relevant for human health and regulatory purposes, including the population-weighted average ozone-season concentration in each region, and the average ozone-season concentration in populated subregions of interest. We further present the monthly distributions of inter-regional transport between source and receptor regions. Finally, we analyze changes in the export and production of ozone to explain differences in the sensitivity of ozone to changes in emissions from different source regions, and whether the transport of ozone itself or of the precursors of ozone is mainly responsible for inter-continental ozone influences.

These changes in emissions of NO_x alone, or NO_x, NMVOCs, and CO together, cause changes in atmospheric CH_4 concentrations through changes in the hydroxyl radical (OH). Methane will respond slowly to changes in OH, and in turn affect global ozone concentrations over the long lifetime of CH_4, including at the surface (Wild et al., 2001; Naik et al., 2005; West et al., 2007). This paper presents only the direct short-term changes in ozone air quality due to emission reductions (i.e., CH_4 concentrations are not allowed to adjust fully to changes in OH); long-term changes in ozone air quality via CH_4 are analyzed in the companion paper to this study (West et al., 2009), as are the consequences for premature human mortality due to exposure to ozone air pollution.

2 Modeling methods

We use the global modeling simulations presented by Naik et al. (2005), and analyze them here for their effects on surface ozone air quality. These simulations use the global chemistry-transport model MOZART-2, and a base simulation that corresponds to the early 1990s (Horowitz et al., 2003). Our simulations have a horizontal resolution of about 2.8° by 2.8°, and 34 vertical levels. The simulations emphasize gas-phase photochemistry and while aerosols are not modeled explicitly, heterogeneous reactions of N_2O_5 and NO_3 on sulfate aerosols are included. Emissions for the early 1990s were compiled by Horowitz et al. (2003) based on several global emissions sources, including EDGAR2.0 (Olivier et al., 1996). The model is driven by meteorological fields

from the middle atmosphere version of the Community Climate Model (MACCM3), which are representative of current atmospheric transport conditions, and do not represent meteorology in a particular year. All simulations are conducted for 25 months, with results presented for the final 12 months.

Anthropogenic emissions of NO_x at ground level are reduced by 10% in each of nine world regions (Fig. 1) individually, as described by Naik et al. (2005). Here, anthropogenic emissions include the combustion of fossil fuels and biofuels (among other sources), but do not include other forms of biomass burning. Reductions of 10% were selected to be small enough to represent the sensitivity to small changes in emissions, avoiding nonlinear chemical responses to large changes in NO_x, while being large enough to distinguish effects on long-range ozone transport (Wild and Akimoto, 2001). This approach differs from other studies that attempt to estimate the total impact of long-range transport by removing emissions entirely (e.g., Berntsen et al., 1999), or by tagging tracers (e.g., Sudo and Akimoto, 2007). Because of the importance of other anthropogenic precursors (NMVOCs, CO, and CH_4) and chemical nonlinearities, our estimates of the sensitivity of long-range ozone transport to 10% regional NO_x reductions may not be good indicators of the total long-range transport among the regions. While Sudo and Akimoto (2007) consider many regions as in this study, they tag ozone based on the region in which the ozone (actually odd oxygen) is formed. In contrast, this study simulates the total effects of small changes in regional emissions, including ozone formed within the region and elsewhere, making a direct comparison with Sudo and Akimoto (2007) difficult. We model idealized controls on emissions of individual pollutants, whereas real actions to control emissions may affect multiple pollutants simultaneously, and may alter the spatial or temporal distributions of emissions. We also analyze simulations by Naik et al. (2005) of simultaneous 10% reductions in anthropogenic emissions of NO_x, NMVOCs and CO from three regions (NA, EU, and SE), to evaluate the effects of changes in NMVOC and CO emissions on surface ozone and long-range transport.

Table 1 presents the anthropogenic NO_x emissions in each region in the base simulation. While our simulations represent emissions from the early 1990s, ozone precursor emissions have likely decreased since that time from NA, EU, and FSU, and are likely to have increased elsewhere (Schultz et al., 2007); increases may be most pronounced in East Asia, where NO_x emissions are likely to have increased by more than 50% (Richter et al., 2005; Zhang et al., 2008). Consequently, we likely underestimate the present-day influences of 10% reductions on long-range transport for rapidly developing regions such as East Asia. Changes in emissions since the early 1990s are likely less important for the changes in ozone per unit change in emissions, although such changes could affect the chemical regime and sensitivity of ozone formation to NO_x and NMVOCs. The uncertainty in our estimates of long-range transport can be estimated by analyzing the spread of several models in the HTAP experiments, which find an average coefficient of variation (σ/μ) of 38% for inter-regional ozone influences when reducing NO_x emissions by 20%, and of 31% when reducing NO_x, NMVOCs, CO, and aerosols by 20% simultaneously (Task Force on Hemispheric Transport of Air Pollution, 2007; Fiore et al., 2009). Similarly, estimates of uncertainty can be derived from studies of the inter-annual variability of transport for particular regions. Duncan and Bey (2004) show little effect of meteorology on annual transport from Europe, and that inter-annual variability in transport is smaller than seasonal variability. For Liu et al. (2005), inter-annual variability is typically 30–40% of average transpacific transport, for passive tracers, with a maximum of 50%. Finally, Li et al. (2002) show large influences of inter-annual variability in monthly average transatlantic transport, associated with transport events, but the effects on annual average transport, the main focus here, are likely smaller.

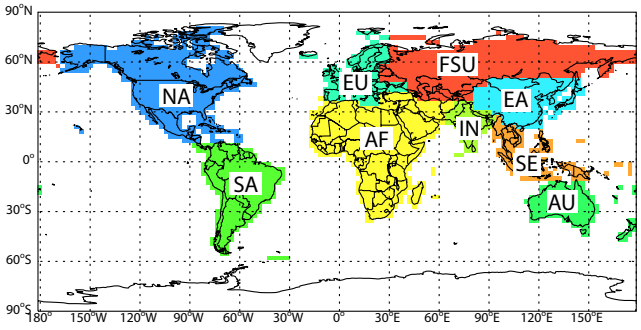

Fig. 1. Definitions for nine world regions.

In Fig. 1, note that the Middle East is included as part of the Africa region, and Central America and the Caribbean are in the North America region. We present changes in surface ozone for several indicators, including the population-weighted ozone concentration. Population-weighting is done by multiplying the concentration and population of each grid cell, summing over all grid cells, and dividing by the total (regional or global) population, where the global distribution of population is taken from the LandScan database (Oak Ridge National Laboratory, 2005) for 2003, and mapped onto the MOZART-2 modeling grid.

3 Model evaluation

Results from the base simulation were compared previously with measurements of ozone and other chemical species aloft, suggesting that model predictions are generally in the range of measured concentrations (Horowitz et al., 2003). In Fig. 2, we compare the base simulation with surface measurements of ozone at non-urban sites from the Clean Air Status and Trends Network (CASTNET) in the

Table 1. Total anthropogenic NO_x emissions, population, and indicators of surface ozone concentration from the base simulation.

Region	Anthropogenic NO_x emissions ($Tg\,N\,yr^{-1}$)	Annual average O_3 (spatially-weighted) (ppb)	Population (million)	Population-weighted annual average O_3 (ppb)	3-month population-weighted average O_3 (ppb)[a]	3-month period with highest O_3^a
NA – North America	8.0	35.2	496.0	35.5	42.6	May – Jul
EU – Europe	4.8	34.9	543.6	32.0	40.4	Jun – Aug
FSU – Former Soviet Union	2.5	31.6	249.1	31.0	39.5	Jun – Aug
AF – Africa	2.0	35.3	1136.4	36.9	39.8	Mar – May
IN – India	1.1	39.2	1382.8	41.0	48.1	Feb – Apr
EA – East Asia	4.0	39.9	1531.9	37.7	43.7	Jul – Sep
SA – South America	0.8	29.1	371.7	29.0	34.2	Aug – Oct
SE – Southeast Asia	0.7	23.2	558.0	28.1	31.2	Dec – Feb
AU – Australia	0.4	21.2	24.7	21.7	25.5	Aug – Oct
Global	24.2	26.0	6302.1	35.9		

[a] The consecutive three-month period with the highest population-weighted ozone concentration in the base simulation.

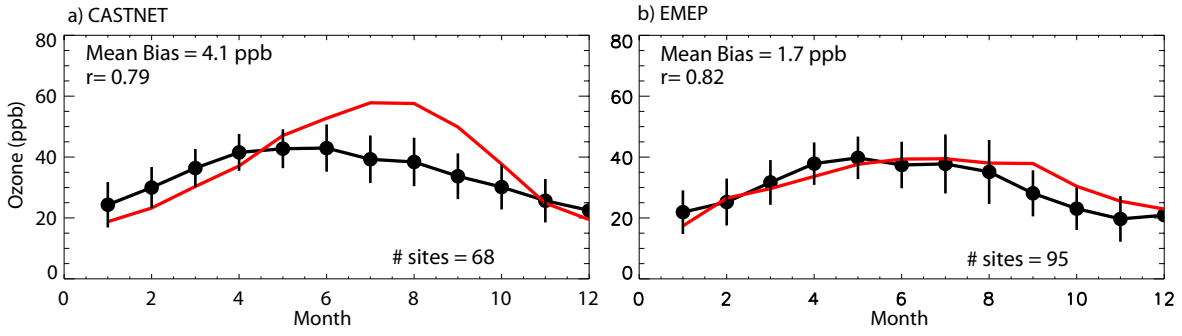

Fig. 2. Comparison of the monthly average modeled (red) and 1990 to 1994 average measured (black) surface ozone concentrations, averaging over **(a)** the CASTNET network in USA, and **(b)** the EMEP network in Europe. Errorbars on the observations show ±1 standard deviation among the multi-year averages for all sites.

USA (www.epa.gov/castnet) and the European Monitoring and Evaluation Programme (EMEP, www.nilu.no/projects/ccc/emepdata.html). Because the simulation uses emissions from the early 1990s with meteorology that does not reflect a particular year, we compare the model prediction with the average of observations between 1990 and 1994. These results show that the model tends to overestimate the annual mean ozone, by 4.1 ppb for CASTNET and 1.7 ppb for EMEP, but that the CASTNET measurements are more substantially overestimated in summer. Analysis of biases at individual stations reveals some regional patterns; for example, the bias is greatest in the Southeastern US and over the Iberian Peninsula. We also compare the model results with measurements at seven remote stations from the NOAA Global Monitoring Division network (www.esrl.noaa.gov/gmd/dv/ftpdata.html), again showing that the model is biased high, by 5.8 ppb for the annual average. However, the bias is particularly large at high altitude stations, and when two such stations are removed (Mauna Loa and Niwot Ridge), the bias for the remaining five stations is 2.8 ppb.

Table 1 shows surface ozone concentrations averaged within each region in the base simulation, for the annual average ozone, and the population-weighted average ozone during the ozone season (the consecutive three-month period with the highest average population-weighted ozone in that region). We analyze peak seasonal concentrations, as these are often of greatest regulatory concern, and by population-weighting the results, we emphasize polluted areas within the receptor region with a metric that is relevant for human health. These results identify IN (India) and EA (East Asia) as the regions with the highest modeled ozone concentrations in the base simulation.

4 Results: short-term changes in ozone air quality

Figure 3 shows the spatial distribution of the change in surface ozone concentrations due to 10% reductions in anthropogenic NO_x emissions in each of the nine source regions, shown for the three-month period with the highest population-weighted ozone in each source region. The

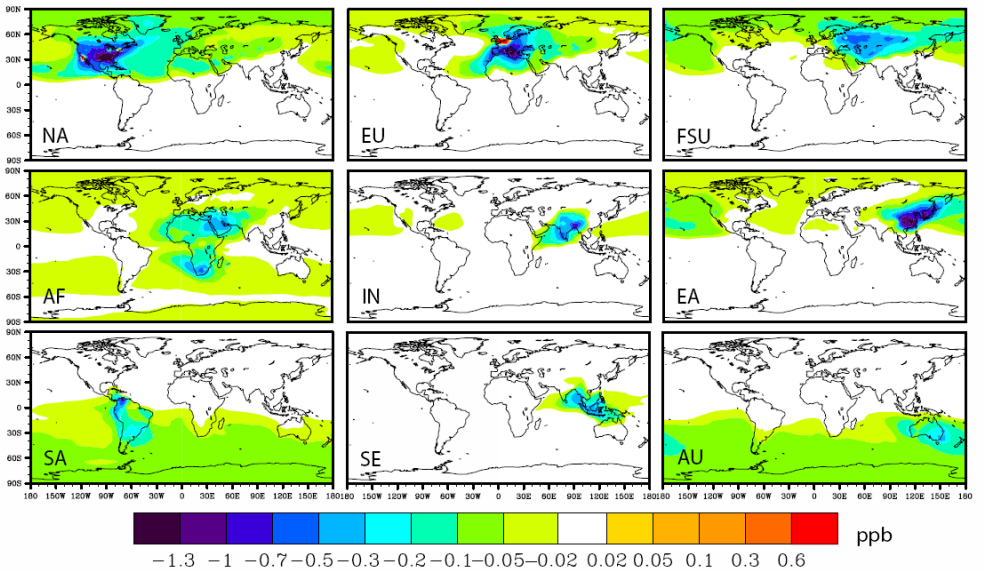

Fig. 3. Change in 3-month average surface ozone concentration due to a 10% reduction in anthropogenic NO_x emissions from each region, shown for the consecutive 3-month period in which the population-weighted ozone is highest in each source region (see Table 1).

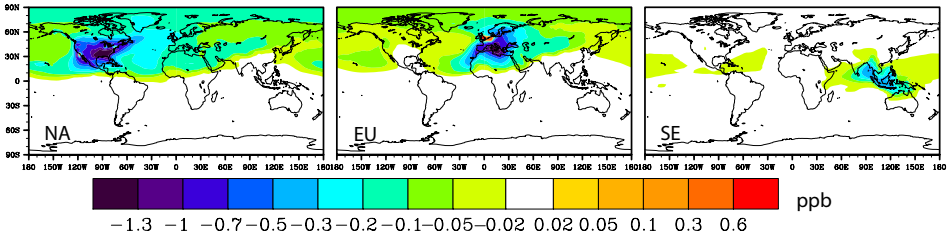

Fig. 4. As Fig. 3, but for the simulations where anthropogenic NO_x, NMVOC, and CO emissions are reduced by 10%.

results typically show surface ozone decreases greater than 0.2 ppb in the source regions themselves, with smaller ozone decreases (>0.02 ppb) observed over areas much larger than the source regions. Reductions >1 ppb are observed within the NA, EU, and EA source regions, with widespread influences throughout the NH, as these regions have the greatest NO_x emissions (and therefore the greatest decrease in NO_x from the 10% reduction). Source regions in the Southern Hemisphere (SH) also have widespread effects on surface ozone in the SH, despite relatively small total NO_x emissions and small effects on ozone within the source regions, in agreement with previous results (Sudo and Akimoto, 2007). Transport between hemispheres is smaller than transport within each hemisphere, given the long interhemispheric mixing time scale (∼1 year). The reductions in NO_x emissions in IN and SE tend to affect surface ozone over a limited spatial extent, while the reductions in AF have widespread effects on ozone, as it has substantial emissions in both hemispheres. Figure 3 also shows that local NO_x reductions cause surface ozone to increase over Northern Europe, Los Angeles, and New York; ozone production in these regions is strongly NO_x-saturated in the model, even in the peak ozone season, such that decreases in NO_x emissions reduce the destruction of ozone by reaction with fresh NO emissions. Other global models also show that some of these regions are NO_x-saturated (Duncan et al., 2008). However, such local sensitivities to changes in emissions are uncertain in a global model, and differ among different global models (Fiore et al., 2009); these sensitivities are best determined using a local or regional model that has been thoroughly tested for local conditions. In addition, there are uncertainties associated with subgrid-scale chemical processing in urban regions, which may affect the export of ozone and its precursors from urban regions to the global model (e.g., Duncan and Chameides, 1998).

Table 2 shows the full source-receptor matrix for nine regions, for population-weighted ozone averaged over the 3-month period with highest ozone in the receptor region. For all source regions, the 10% reduction in anthropogenic NO_x

Table 2. Source-receptor matrix of the change in 3-month population-weighted average ozone concentration (parts per trillion, ppt) in each receptor region due to 10% NO_x emission reductions in each source region (the 3-month period is defined for each receptor region in Table 1). Values underlined indicate the effect of a reduction in one region on itself (the diagonal). The nine highest inter-regional values are in bold.

		Receptor Region								
		NA	EU	FSU	AF	IN	EA	SA	SE	AU
Source Region	NA	−512	**−63**	**−46**	**−44**	**−44**	−8	−11	−10	−3
	EU	−8	−194	**−184**	**−73**	−13	−9	1	−3	1
	FSU	−14	**−55**	−401	−16	−16	−27	0	0	0
	AF	−4	−9	−15	−176	**−50**	−1	−8	−8	−17
	IN	−4	0	−2	−6	−482	−16	−2	−32	−1
	EA	−16	−7	−16	−6	−6	−930	−1	**−105**	0
	SA	−6	1	1	−4	−4	1	−252	−5	−34
	SE	0	2	1	−2	−15	−38	−9	−265	−11
	AU	0	0	0	−1	0	0	−13	−3	−179

emissions has the greatest impact on ozone within that region. The largest inter-regional influences identified are from EU to FSU, EA to SE, and EU to AF, as these regions are contiguous. With these exceptions, intra-regional influences are generally larger than inter-regional influences by a factor of ten or more. The largest impacts, either within a source region or between regions, often result from the source regions NA, EU, and EA, as these have the largest NO_x emissions.

While Table 2 highlights the regions with highest emissions, the regions with the greatest effects on ozone per unit change in NO_x emissions are often in the tropics and SH (Table 3). The regions AU, IN, and SE have the largest effects per ton within their own regions, and some of the largest inter-regional influences are between regions in the tropics, in particular from SE to EA and SA to AU. The reasons for this enhanced sensitivity in the tropics and SH are explored in Sect. 4.3.

In Table 4 and Fig. 4, we show the changes in surface ozone for the three simulations where regional anthropogenic emissions of NO_x, NMVOCs, and CO are reduced by 10% simultaneously. For the NA and EU reductions, the magnitudes of the surface ozone responses for both intra- and inter-regional influences, are comparable to those of Wild and Akimoto (2001). Comparing with Table 2, we see that adding the NMVOC and CO reductions causes only small additional decreases in ozone within the source regions NA and SE. However, reducing NMVOCs and CO more than doubles the ozone decrease for EU on itself, showing that the ozone formation in EU is strongly NO_x-saturated in the model. For all source-receptor pairs considered, reducing all three precursors decreases ozone more than when only NO_x is reduced, with the ratio of results in Table 4 to Table 2 ranging from 1.3 to 3.0 (omitting the source-receptor pairs with decreases <10 ppt in Table 2). Reducing all three precursors in NA causes a greater relative influence on the receptor regions it influences most (1.7 to 1.9 times the results in Table 2 for the EU, FSU, AF, and IN receptor regions) than its effect on itself (1.3 times that in Table 2). Likewise, the ratio when reducing all three precursors in the SE source region is greater for the receptors it influences most (1.3 to 1.7 for IN, EA, and AU) than its effect on itself (1.1). These results suggest that the changes in emissions of NMVOCs and CO have important influences on long-range ozone transport, which will be analyzed further in Sect. 4.3.

We also calculate the source-receptor relationships for the annual average spatially-weighted change in ozone, a metric that would be used commonly in the atmospheric science literature. Comparing Tables and S1 and S2, see http://www.atmos-chem-phys.net/9/6077/2009/acp-9-6077-2009-supplement.pdf, with Tables 2 and 4, respectively, the strongest source-receptor pairs are generally the same, but there are important quantitative differences. This suggests that the quantification of long-range transport can vary with the ozone concentration metric used. We likewise compare our results with the multi-model ensemble results of the HTAP model intercomparison exercise (Table S3), by using the same four receptor regions as defined by Fiore et al. (2009) and the same annual average spatially-weighted indicators. In our case, however, the source regions (Fig. 1) differ from those of Fiore et al. (2009). In general, the inter-continental influences are smaller than the HTAP multi-model mean, indicating a lower sensitivity in our modeling; this is also the case in Table S4, where we normalize for the change in NO_x emissions, and Table S5 for simultaneous NO_x, NMVOC, and CO reductions. In addition to different source region definitions, this study used different meteorology and emissions for the early 1990s rather than for 2001. In the HTAP model intercomparison, with all models simulating 2001 conditions, MOZART-2 also shows a relatively low sensitivity of ozone and its long-range transport to changes in emissions.

Table 3. Normalized source-receptor matrix showing the change in 3-month population-weighted average ozone (Table 2) per unit change in NO_x emissions (ppb $(TgN\,yr^{-1})^{-1}$). Note that negative ozone changes divided by the negative change in NO_x emissions gives mainly positive values here.

		Receptor Region								
		NA	EU	FSU	AF	IN	EA	SA	SE	AU
Source Region	NA	0.64	0.08	0.06	0.06	0.05	0.01	0.01	0.01	0.00
	EU	0.02	0.40	**0.38**	0.15	0.03	0.02	0.00	0.01	0.00
	FSU	0.06	**0.22**	1.62	0.07	0.06	0.11	0.00	0.00	0.00
	AF	0.02	0.04	0.07	0.89	**0.25**	0.00	0.04	0.04	0.08
	IN	0.04	0.00	0.02	0.05	4.19	0.14	0.02	**0.28**	0.01
	EA	0.04	0.02	0.04	0.02	0.02	2.33	0.00	**0.26**	0.00
	SA	0.07	−0.02	−0.01	0.05	0.05	−0.01	3.07	0.06	**0.41**
	SE	0.00	−0.02	−0.01	0.04	**0.23**	**0.58**	0.13	4.05	0.16
	AU	−0.01	−0.01	−0.01	0.02	0.02	−0.01	**0.34**	0.08	4.70

Table 4. Source-receptor matrix for the simulations where anthropogenic emissions of NO_x, NMVOCs, and CO are all reduced by 10% (for the 3-month population-weighted average ozone, in ppt).

		Receptor Region								
		NA	EU	FSU	AF	IN	EA	SA	SE	AU
Source Region	NA	−668	**−113**	**−77**	**−86**	**−80**	−17	−16	−29	−6
	EU	−29	−407	**−240**	**−134**	−40	−18	−2	−21	−1
	SE	−4	−1	−2	−8	−25	−47	−15	−300	−17

4.1 Seasonal cycle of changes in long-range ozone transport

In Fig. 5, we analyze the monthly surface ozone response to the regional 10% NO_x reductions averaged within each receptor region. Considering NA as a receptor, the 10% NO_x reduction in EA causes the greatest reduction in ozone in October and November, with another peak in April, but has a substantially smaller influence during the summer when ozone is highest in NA. This seasonal distribution of transpacific transport is well documented, although other studies that used different meteorology have identified spring as the period of greatest transport (Wild and Akimoto, 2001; Liu et al., 2005). Similarly, the influence of NA on EU is maximum in spring and fall, in agreement with previous results (Li et al., 2002). AF is substantially influenced by the NO_x reductions in EU and FSU during April through August, while the influence of NA is relatively uniform throughout the year.

In IN during the period of highest ozone (February through April), ozone responds most strongly to the NO_x reduction in NA and AF. During the monsoon season of June through September, long-range influences on ozone in IN are small. Following the monsoon season, however, SE and EA have substantial influences on ozone in IN, as winds during these months are predominantly from the Northeast and East. SE likewise experiences the greatest influence from EA during this period. EA is influenced most strongly, during its period of highest ozone, by the NO_x reductions in SE and FSU, while the influences of NA, EU, and AF are higher in other seasons.

Figure 6 shows the monthly effects of NO_x reductions on ozone in the source regions themselves, with influences that are typically much larger than the inter-regional influences in Fig. 5. Figure 6 shows a clear summer maximum in the influence of NO_x reductions within the source regions in the temperate NH (NA, EU, FSU, and EA). While ozone decreases within these source regions in the NH summer, it increases in the NH winter due to local titration with NO emissions, in agreement with other model results (Wild and Akimoto, 2001; Duncan et al., 2008). This winter increase in ozone is often weaker when considering inter-regional transport (Fig. 5). The NO_x reductions in EU cause a strong increase in ozone in the FSU in winter, but relatively smaller increases in winter in NA, AF, and EA.

Despite the fact that NO_x reductions in NA cause the greatest within-region reductions in summer (Fig. 6), its influences on EU and FSU are greater in the NH spring and fall, and the influences on AF, IN, EA, and SA are actually greatest in the NH winter. This could be due to conditions more conducive to transport outside of the NH summer, as well as the longer lifetime of ozone in the colder seasons. Likewise, the NO_x reductions in EA have large influences on ozone in NA, EU, IN, and SE in October and November, after the period of the greatest influence within

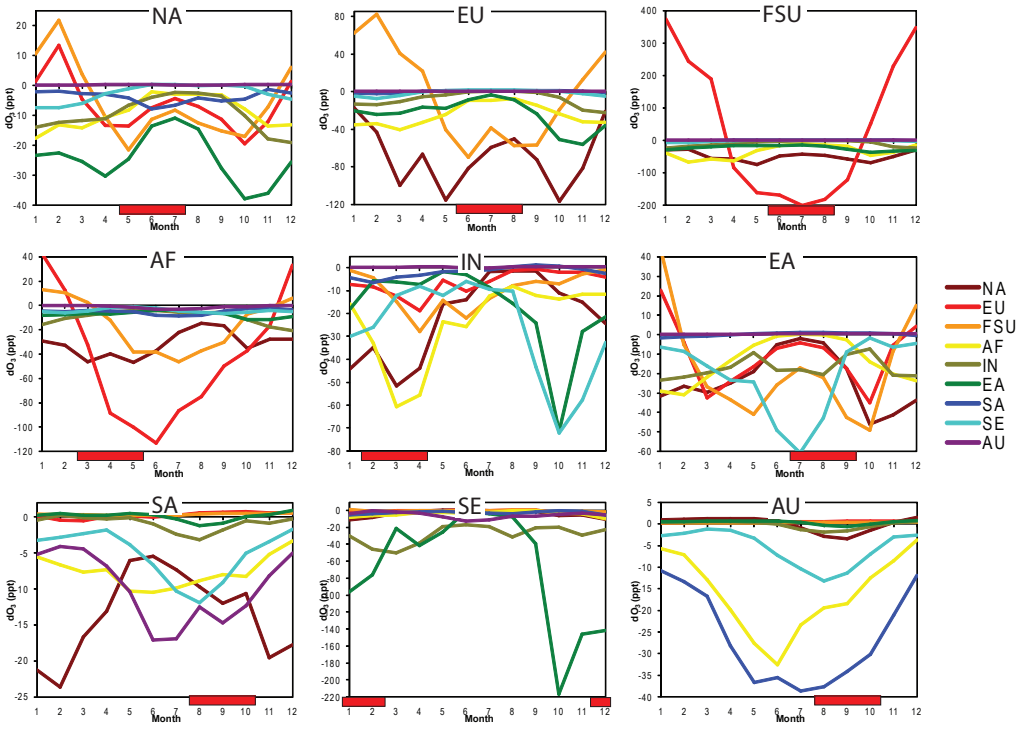

Fig. 5. Monthly influences of 10% regional NO_x reductions, where each panel depicts the influences of several source regions on a receptor region, for the monthly-average population-weighted ozone concentration. The effects of source regions on themselves are omitted (see Fig. 6), as these are typically much larger than the inter-regional influences. Red bars on the horizontal axis indicate the three-month period when the modeled population-weighted average ozone is highest in each receptor region. Note that the vertical axes differ in each panel.

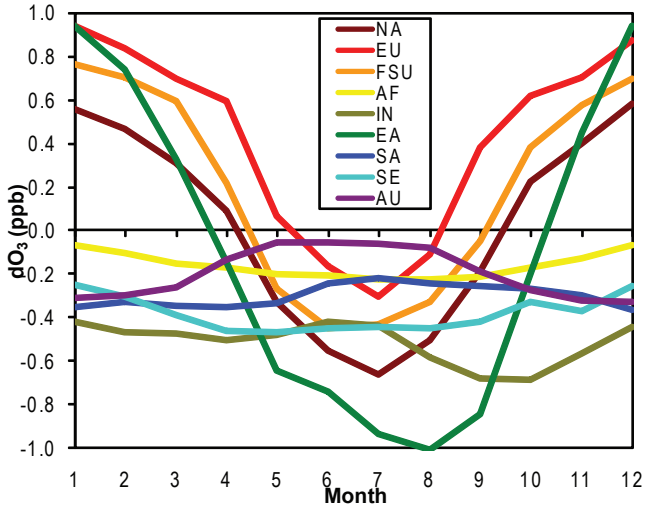

Fig. 6. Monthly influences of 10% regional NO_x reductions on ozone in the source regions themselves, for the monthly-average population-weighted ozone concentration.

EA. This is also the period in which EA is most influenced by NO_x reductions from other source regions in the NH (NA, EU, FSU), suggesting that the meteorological conditions that favor long-range transport from EA, including less stagnation and greater vertical mixing, also cause greater import of ozone from elsewhere in the NH. For the source regions NA and EA, as well as for EU and FSU for some receptor regions, these results show that while the greatest changes in ozone production occur in the summer, the conditions for transport to other NH receptors are more prevalent at other times of year.

These monthly influences are quantified in Table S6. While Table 2 shows the influence on ozone during the three-month period when ozone is highest at the receptor, Table S6 shows the maximum three-month average influence for each source-receptor pair. Here, for example, the influence of the NO_x reduction in EA on several receptor regions in the NH is substantially greater than shown in Table 2, because the period of maximum influence does not coincide with the period of highest ozone in the receptor region.

4.2 Effects of NO_x reductions on populated subregions of interest

In addition to evaluating the impacts of regional NO_x reductions on ozone over continental regions, we can also consider effects using smaller subregions as receptors. Table 5 shows the changes in the three-month average ozone in selected subregions. We select subregions that have high population densities, significant ozone pollution, or lie close to the border of different regions to illustrate inter-regional transport. We define large subregions using 3 by 3 grid cells (roughly 8.4° by 8.4°, see Fig. S1), larger than any single metropolitan area, so that the results are not strongly influenced by local chemistry in the urban region, which is uncertain in the global model, while inter-continental influences are captured.

In Table 5, the NO_x reduction in EA has a greater influence on Los Angeles than on Toronto or the population-weighted average for NA (Table 2). Likewise, the NO_x reduction in NA has a stronger influence on London than populated subregions further east. Reductions in NO_x from EU increase ozone in London, as ozone formation in this region is strongly NO_x-saturated in the model (Fig. 3). Athens, Moscow, Cairo, and Tehran are all influenced strongly by NO_x reductions in the regions EU, FSU, and AF, illustrating the strong influences that these regions have on one another. Bangkok and Manila are strongly influenced by EA, although Jakarta is not, showing the lack of transport across the equator. For subregions in IN, reductions in NO_x emissions in SE and EA strongly reduce ozone in Chennai, as these regions mainly affect ozone in Southern India. Delhi, on the other hand, shows very little influence from emissions from SE, but is influenced more by the reductions in AF and NA.

If we normalize the influences on ozone per unit change in NO_x emissions, as in Table 3 (see Table S7), we see that NO_x reductions from EA are on average about 20% as effective per ton at improving ozone air quality in Los Angeles, as reductions in NA. Likewise, NO_x reductions from EU are on average about 20% as effective per ton at improving ozone air quality in Cairo, as reductions in AF. For this comparison, we compare the effects of 10% NO_x emission reductions over the large continental regions. Had we assessed the effect of NO_x reductions only in the Cairo area, for example, they would presumably be more effective per ton at improving ozone in Cairo than reductions that take place over all of AF (but depending on the local chemical response), and the effectiveness of NO_x reductions in EU would be less than 20% as effective as reductions in the Cairo area itself.

Table S8 shows these effects on subregions for the simulations in which NO_x, NMVOCs, and CO are reduced simultaneously. For all source-receptor influences shown, the results are greater (more negative) than in Table 5, in agreement with the relationships between Tables 2 and 4.

4.3 Changes in ozone production and export

In Table 3, surface ozone is more sensitive to changes in emissions from tropical and SH regions than from temperate NH regions. Here we analyze changes in the total production and export of ozone and precursors to explain this enhanced sensitivity. We further use this analysis to assess whether the transport of ozone or of its precursors is more important for inter-continental ozone transport.

Consistent with Table 3, Table 6 shows that the change in the tropospheric ozone burden (defined as below the "chemical tropopause" of 150 ppb ozone in the base simulation) per unit change in NO_x emissions is greatest for NO_x reductions from SE, SA, and AU. The large difference in sensitivity between temperate regions and those in the tropics and SH agrees with previous results (Fuglestvedt et al., 1999; Berntsen et al., 2005; Naik et al., 2005).

One explanation for the greater sensitivity in tropical source regions is the year-round photochemical activity in the tropics. In addition, global emissions of NO_x are strongly concentrated in the industrialized source regions of the temperate NH, with 80% of global anthropogenic emissions in NA, EU, FSU, and EA (Table 1). With a much lower density of NO_x emissions in tropical regions and the SH, ozone concentrations would be expected to be more sensitive to changes in NO_x emissions, due to the nonlinearity of ozone chemistry with respect to its precursors. Furthermore, active convection in tropical regions can transport ozone and its precursors more effectively to the upper troposphere, thereby increasing their lifetimes and the ability of precursors to produce ozone. While greater water vapor concentrations (causing greater loss of ozone by reactions of $O(^1D)+H_2O$, O_3+OH, and O_3+HO_2) and convective mixing (bringing ozone into contact with vegetated surfaces) can decrease the ozone lifetime in the tropics (Lawrence et al., 2003), these processes are clearly not dominant in the model since Table 6 shows greater sensitivity.

In Table 6, the decrease in regional NO_x emissions in each region decreases ozone production, and increases the global average ozone lifetime. Because the global ozone burden decreases for all source regions, we can infer that the change in production dominates over the increase in lifetime, which would tend to increase ozone. In Table 6, the greater change in ozone burden per unit change in NO_x emissions for regions in the tropics and SH is partially explained by the greater change in ozone production per unit emissions, with SE having the greatest change for both quantities. Table 6 also shows that SE has by far the highest fraction of the change in ozone production (35%) occurring in the upper troposphere (above 469 mb), with high values for other regions near the equator (IN and SA), suggesting greater vertical transport of precursors. Likewise, the ozone lifetime diagnosed from the change in regional emissions is greatest from tropical and SH regions, perhaps due partly to the greater fraction of ozone production in the upper troposphere. Even for SE, however,

Table 5. Change in 3-month average ozone (ppt) in selected receptor subregions due to 10% NO_x reductions from source regions. Ozone changes are area-weighted within the 8.4° × 8.4° receptor regions, and reported for the three months with highest ozone in the base simulation (in parenthesis). See supporting information for region definitions and results per unit emission reduction.

		Receptor Region												
		Los Angeles (JJA)	Mexico City (OND)	Toronto (JJA)	London (MAM)	Rome (JJA)	Athens[a] (JAS)	Moscow (JJA)	Tashkent (JJA)	Lagos (JFM)	Johannesburg (SON)	Cairo (MJJ)	Nairobi (DJF)	Tehran[b] (MJJ)
Source Region	NA	−411	−623	−572	**−123**	−51	−49	−39	−76	−15	0	−58	−4	−82
	EU	−12	−7	−12	801	−827	−656	**−254**	**−102**	−13	1	**−262**	0	**−112**
	FSU	−23	−3	−23	0	−20	−87	−393	−416	−1	1	−35	1	**−323**
	AF	−1	−9	0	−24	−10	**−135**	−1	−18	−130	−189	−512	−74	−198
	IN	−8	−22	−1	−8	0	−2	0	−10	−1	0	−4	−42	−6
	EA	−46	−29	−17	−26	−6	−13	−8	−33	0	1	−8	1	−13
	SA	0	−3	1	0	1	1	1	1	−5	−22	1	−4	0
	SE	1	−4	1	−2	2	1	1	1	−3	−4	1	−10	0
	AU	1	1	0	0	0	1	0	0	0	−12	1	0	0

Table 5. Continued.

		Receptor Region												
		Dubai (AMJ)	Delhi (MAM)	Chennai (DJF)	Dhaka (FMA)	Hong Kong (OND)	Beijing (JAS)	Tokyo (FMA)	Caracas (MAM)	Sao Paulo (ASO)	Bangkok (DJF)	Jakarta (JAS)	Manila (DJF)	Sydney (ASO)
Source Region	NA	−71	−71	−15	−25	−28	−17	−62	−95	−2	−14	−3	−33	−2
	EU	−65	−24	−1	−3	−14	−20	−41	−4	1	−5	1	−10	1
	FSU	−67	−38	0	−2	−10	−62	−27	0	0	−1	1	−1	0
	AF	−356	−83	−5	−19	−14	−1	−40	−10	−8	−7	−3	−17	−17
	IN	−27	−363	−319	−763	−25	−7	−15	−4	−1	−21	−2	−37	−1
	EA	−10	−10	−45	−5	−583	−810	177	−3	0	**−232**	−1	**−134**	0
	SA	−2	−3	−7	−4	0	1	−1	−199	−215	−3	−12	−6	−32
	SE	−2	−5	**−106**	−34	−7	−5	−6	−4	−8	−243	−228	−89	−10
	AU	0	0	0	0	1	0	0	0	−23	0	−26	0	−300

[a] The region defined for Athens lies mainly in EU, and partly in AF.
[b] The region defined for Tehran lies mainly in AF, and partly in FSU.

the majority of the change in ozone production occurs in the lower troposphere. All regions also have a majority of the change in ozone production below the top of the mixing layer (taken as 744 mb), except for SE (with 52% above).

In Table 7, we analyze changes in the tropospheric NO_y burden as an indicator of the transport of NO_x and its oxidation products that can lead to ozone formation. The tropical and SH regions have the greatest change in NO_y burden per unit change in emissions, leading to large decreases in ozone production. The fraction of the change in NO_y burden in the upper troposphere is also greatest for these same regions, as is the NO_y lifetime diagnosed from the change in emissions. The greater vertical convection for tropical regions is therefore important in transporting ozone and NO_y to the upper troposphere, which is expected to contribute to

Table 6. Changes in global annual mean tropospheric O_3 burden (B_{O3}), global annual O_3 production (P_{O3}) and net export from the source region (X_{O3}), also shown per unit change in NO_x emissions (E_{NOx}), the O_3 lifetime, and fractions of these quantities above the source region and in the upper troposphere.

	ΔB_{O3} (TgO_3)	$\Delta B_{O3}/\Delta E_{NOx}$ (TgO_3 $(TgN\,yr^{-1})^{-1}$)	ΔP_{O3} ($TgO_3\,yr^{-1}$)	$\Delta P_{O3}/\Delta E_{NOx}$ ($TgO_3\,yr^{-1}$ $(TgN\,yr^{-1})^{-1}$)	Regional O_3 lifetime ($\Delta B_{O3}/\Delta P_{O3}$) (days)[a]	Global ΔO_3 lifetime (B/P) (days)[b]	Global ΔO_3 lifetime/ ΔE_{NOx} (days $(TgN\,yr^{-1})^{-1}$)
NA	−0.464	0.58	−14.50	18.2	11.7	0.037	−0.046
EU	−0.091	0.19	−4.77	9.9	7.0	0.017	−0.034
FSU	−0.069	0.28	−3.62	14.7	7.0	0.013	−0.051
AF	−0.241	1.21	−7.32	36.9	12.0	0.018	−0.090
IN	−0.193	1.68	−4.69	40.9	15.0	0.009	−0.076
EA	−0.247	0.62	−7.52	18.9	12.0	0.019	−0.047
SA	−0.266	3.25	−6.43	78.5	15.1	0.012	−0.144
SE	−0.304	4.65	−5.93	90.6	18.7	0.007	−0.100
AU	−0.120	3.14	−2.55	66.7	17.2	0.004	−0.094

Table 6. Continued.

	Fraction of global ΔB_{O3} above source region	Fraction of global ΔP_{O3} above source region	Fraction of global ΔP_{O3} above 469 mb	ΔX_{O3} from source region ($TgO_3\,yr^{-1}$)	ΔP_{O3} outside of source region ($TgO_3\,yr^{-1}$)
NA	0.205	0.755	0.155	−3.51	−3.55
EU	0.106	0.631	0.068	−0.73	−1.76
FSU	0.259	0.799	0.041	−0.82	−0.73
AF	0.255	0.870	0.102	−2.16	−0.95
IN	0.112	0.803	0.213	−1.85	−0.93
EA	0.157	0.809	0.112	−2.25	−1.43
SA	0.177	0.749	0.189	−1.92	−1.61
SE	0.140	0.799	0.355	−2.39	−1.19
AU	0.076	0.511	0.086	−0.54	−1.25

[a] We use $\Delta B_{O3}/\Delta P_{O3}$ to diagnose the ozone lifetime associated with changes in regional NO_x emissions, assuming it is not changed significantly by the small NO_x emission change.
[b] The global ozone lifetime (B/P) is calculated to be 24.19 days in the base simulation, and changes in this lifetime due to regional NO_x emission changes are reported.

a longer lifetime of ozone and enhance the ozone production per unit change in NO_x emissions.

Because most of the change in ozone production occurs in the lower troposphere, we conclude that the enhanced ozone sensitivity per unit NO_x emissions for tropical and SH regions is mainly due to greater sensitivity to NO_x in the lower troposphere, and only partially due to the greater vertical transport. For surface ozone air quality, production in the lower troposphere is expected to be even more important than for the tropospheric burden. This result is clarified by analyzing AU, the only region entirely in the SH. While AU has a large change in ozone burden per unit change in NO_x emissions, the fractions of the changes in ozone production and NO_y burden occurring in the upper troposphere are relatively small, as the majority of AU emissions are south of the tropical region of high convective activity. The large sensitivity to changes in NO_x for AU is therefore due to the relative lack of NO_x and greater sensitivity of ozone production to NO_x in the SH, mainly in the lower troposphere. Note that while much of AF is in the tropics, the majority of NO_x emissions are outside of the tropics in the Middle East or in South Africa, where less is transported to the upper troposphere.

Previous studies have emphasized that while vertical transport is not very important for the export of pollution from North America and Europe annually, it is important during the summer (Li et al., 2002; Duncan and Bey, 2004), and especially for transport from East Asia in the seasons

Table 7. Changes in global annual average NO_y burdens, globally and outside of the source region, also shown per unit change in NO_x emissions.

	ΔB_{NOy} (GgN)	$\Delta B_{NOy}/\Delta E_{NOx}$ (GgN (TgN yr^{-1})$^{-1}$)[a]	ΔB_{NOy} outside of source region (GgN)	$\Delta B_{NOy}/\Delta E_{NOx}$ outside of source region (GgN (TgN yr^{-1})$^{-1}$)	Fraction of ΔB_{NOy} outside of source region	Fraction of ΔB_{NOy} above 469 mb
NA	−5.93	7.43 (2.7)	−2.29	2.87	0.39	0.141
EU	−3.59	7.48 (2.7)	−1.88	3.92	0.52	0.043
FSU	−2.16	8.78 (3.2)	−0.70	2.84	0.32	0.041
AF	−1.65	8.30 (2.2)	−0.63	3.19	0.38	0.179
IN	−1.06	9.27 (3.4)	−0.68	5.90	0.64	0.394
EA	−2.39	6.00 (2.2)	−1.03	2.58	0.43	0.197
SA	−1.08	13.20 (4.8)	−0.66	8.02	0.61	0.397
SE	−0.86	13.19 (4.8)	−0.59	9.08	0.69	0.615
AU	−0.35	9.22 (3.4)	−0.24	6.20	0.67	0.177

[a] The $\Delta B_{NOy}/\Delta E_{NOx}$ is expressed as a lifetime of NO_y in parenthesis (days) associated with the regional change in NO_x emissions, assuming the lifetime is not changed significantly by the small NO_x emission change.

of greatest transpacific transport (Wild and Akimoto, 2001; Liu et al., 2003; Sudo and Akimoto, 2007). In Tables 6 and 7, a larger fraction of the NO_y burden change occurs in the upper troposphere for EA than for NA and EU, but the fraction of the ozone production change in the upper troposphere is greater for NA than EA. More importantly, Tables 6 and 7 suggest that vertical pollutant transport will be relatively more important for tropical source regions (SE, IN, and SA), for which inter-continental air pollutant transport has received less attention in the literature.

Table 6 also shows that, for all source regions, a majority of the change in ozone production occurs over the source region itself, although most of the change in global ozone burden is outside of the source region. For most regions, the decrease in the net export of ozone is greater than the decrease in ozone production outside of the source region (last two columns of Table 6), suggesting that while the export of NO_y contributes to long-range ozone transport, the direct export of ozone formed over the source region itself is more important. However, the change in production outside of the source region is greater than the change in export for three regions, EU, NA, and AU. For EU, and to a lesser extent NA, the NO_x reductions in winter cause ozone to increase over the source region, while NO_y is exported and can form ozone elsewhere. The annual average decrease in export is less than the decrease in production outside of the source region, although this result may differ in the summer. For AU, the source region is small and a lack of NMVOCs may contribute to low ozone production over the source region itself. Table 7 shows that for the reduction in AU, a majority of the global change in NO_y burden occurs outside of the source region, and with greater sensitivity to NO_x in the SH, this causes the change in production of ozone outside of the source region to exceed the change in ozone export.

Figure 7 shows the change in the global tropospheric NO_y burden associated with the decreases in regional NO_x emissions. While the change in ozone burden occurs mainly outside of the source region, the fractional change in NO_y burden outside of the source region varies between 32% and 69% among the different regions, with relatively more of the change in NO_y burden occurring outside of the source regions in the tropics and SH (Table 7).

When NMVOC and CO emissions are reduced in addition to NO_x, the decreases in ozone burden and production are greater than when NO_x is reduced alone, with a larger relative response for the reduction in EU (Table 8), in agreement with the changes for surface ozone (comparing Table 4 to Table 2). Relative to Table 6, the NMVOC and CO reductions cause a greater decrease in ozone exported from the source regions. However, the change in ozone production outside of the source region, relative to Table 6, is much larger than the change in export, and in fact, most of the global change in ozone production is accounted for by the change in ozone production outside of the source region. When reducing all precursors, we see that the decrease in ozone production outside of the source region is now greater than the decrease in export from the source region for all three regions. In agreement, Table 8 shows that the fraction of the change in ozone burden and production above the source region is smaller than in Table 8, while the fraction of ozone production in the upper troposphere is greater.

In Table 9, reducing NMVOC and CO emissions in addition to NO_x causes a greater decrease in the global NO_y burden, with much of this additional decrease (relative to Table 7) occurring outside of the source region. This can be explained in part by the importance of NVMOCs for producing PANs. By reducing NMVOCs, less of the NO_x is converted to PANs and more to nitric acid (HNO_3). Since the lifetime of PANs is greater than NO_x or HNO_3, the mean lifetime

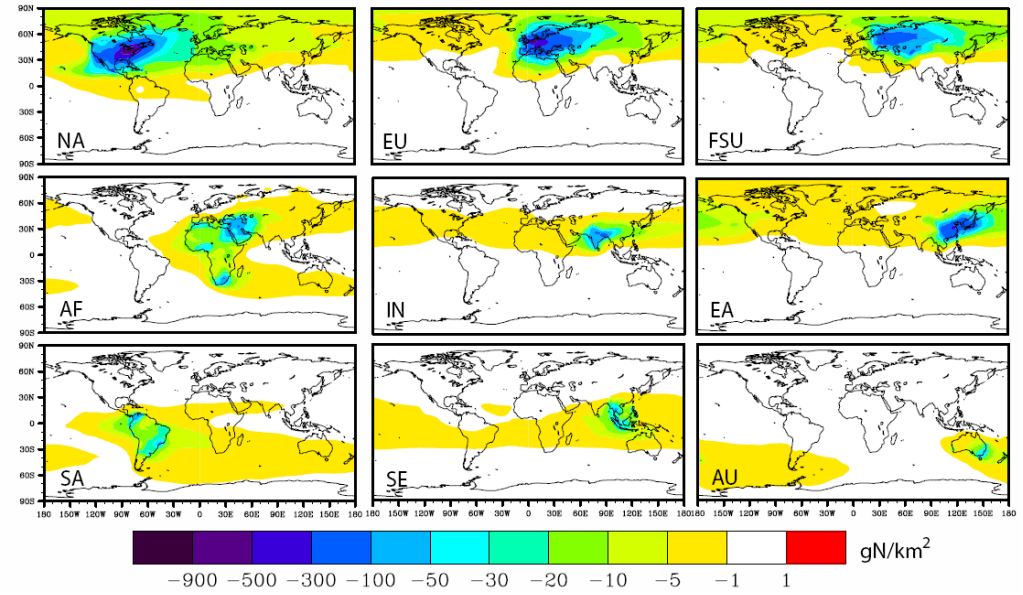

Fig. 7. Change in the annual average tropospheric burden of NO_y due to 10% reductions of anthropogenic NO_x emissions from each region.

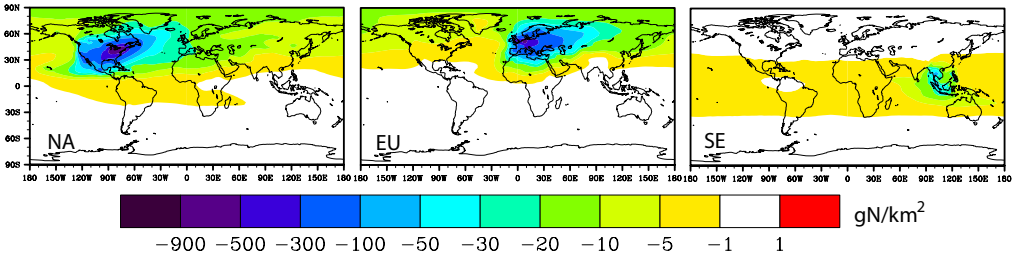

Fig. 8. As Fig. 7 but for the simulations where anthropogenic NO_x, NMVOC, and CO emissions are reduced by 10%.

of NO_y decreases, as does its burden. Additionally, CO decreases can cause OH to increase, leading to faster conversion of NO_x to HNO_3, which also shortens the NO_y lifetime. Accordingly, the fraction of the change in NO_y burden outside of the source region is greater in Table 9 than in Table 7. Likewise, Fig. 8 shows a more widespread change in NO_y burden than in Fig. 7.

These findings together suggest that the decreases in NMVOC and CO emissions are important for long-range ozone transport, and have perhaps been underemphasized in past analyses of long-range transport. Likewise, Fiore et al. (2009) find that the ratio of impacts from foreign sources relative to domestic sources is greater for changes in NMVOCs and CO emissions than for NO_x. We do not attempt to distinguish the relative importance of NMVOCs and CO for long-range transport, but Fiore et al. (2009) find that a 20% decrease in anthropogenic NMVOC emissions causes greater impacts on both intra- and inter-regional ozone than does a 20% decease in anthropogenic CO.

5 Conclusions

Here we analyze the effects of 10% reductions in anthropogenic emissions of NO_x from nine world regions on the long-range transport of ozone. The results identify the source-receptor pairs for which the long-range transport influence is greatest: EU on FSU (comparable to the influence within EU itself), EA on SE, and EU on AF, for three-month population-weighted ozone metrics. Most other inter-regional influences are roughly a factor of 10 smaller than the ozone influence within the source region. By normalizing these results per unit of NO_x reduced, we consider the relative efficacy of emission controls in different regions. For example, we find that NO_x reductions in NA are ∼20% as effective at improving ozone air quality in EU as reductions in EU itself. For NO_x reductions in NA to be cost-effective at reducing ozone in EU (without considering the benefits of ozone reductions in NA) they would have to cost less than 20% per ton of the cost of NO_x reductions in EU.

Table 8. As Table 6, for the simulations where NO_x, NMVOCs, and CO are reduced simultaneously ("ratio to reducing only NO_x" compares to results in Table 6).

	Anthrop. CO emissions ($Tg\,yr^{-1}$)	Anthrop. NVMOC emissions ($TgC\,yr^{-1}$)	ΔB_{O3} (TgO_3)	ΔB_{O3} ratio to reducing only NO_x	ΔP_{O3} ($TgO_3\,yr^{-1}$)	ΔP_{O3} ratio to reducing only NO_x	Regional O_3 lifetime ($\Delta B_{O3}/\Delta P_{O3}$) (days)	Global ΔO_3 lifetime (B/P) (days)	Fraction of global ΔB_{O3} above source region	Fraction of global ΔP_{O3} above source region	Fraction of global ΔP_{O3} above 469 mb	ΔX_{O3} from source region ($TgO_3\,yr^{-1}$)	ΔP_{O3} outside of source region ($TgO_3\,yr^{-1}$)
NA	100.0	4.7	−0.653	1.41	−18.77	1.29	12.7	0.044	0.185	0.655	0.163	−4.06	−6.47
EU	69.3	3.5	−0.215	2.36	−7.77	1.63	10.1	0.022	0.078	0.476	0.096	−1.11	−4.07
SE	41.2	2.6	−0.421	1.38	−7.92	1.34	19.4	0.008	0.116	0.643	0.372	−2.58	−2.83

Table 9. As Table 7, for the simulations where NO_x, NMVOCs, and CO are reduced simultaneously ("ratio to reducing only NO_x" compares to results in Table 7).

	ΔB_{NOy} (GgN)	ΔB_{NOy} ratio to reducing only NO_x	ΔB_{NOy} outside of source region (GgN)	ΔB_{NOy} outside of source region ratio to reducing only NO_x	Fraction of ΔB_{NOy} outside of source region	Fraction of ΔB_{NOy} above 469 mb
NA	−6.51	1.10	−2.79	1.22	0.43	0.159
EU	−4.08	1.14	−2.35	1.25	0.58	0.065
SE	−1.13	1.31	−0.84	1.42	0.74	0.608

We similarly analyze the long-range transport influences on particular subregion receptors globally, showing that long-range influences can differ substantially among metropolitan areas in the same continental regions. The influences of 10% regional NO_x reductions on long-range ozone transport are shown to vary strongly with season, where the seasons of greatest long-range transport often do not coincide with the seasons when ozone is highest in the receptor regions.

We find that the change in tropospheric ozone burden, and its long-range transport, per unit reduction in NO_x emissions, is largest for source regions in the tropics and SH. We attribute this high sensitivity to a relative lack of NO_x in these regions, as well as greater photochemical activity, causing greater sensitivity of ozone production to changes in NO_x in the lower troposphere. For tropical regions, the increased vertical transport of ozone and its precursors, due to greater convection, is a secondary cause of this enhanced sensitivity. For most source regions, the export of ozone from the source region is relatively more important for long-range transport than the reduced export of NO_y and reduced production of ozone outside of the source region.

Reductions in emissions of NMVOCs and CO also influence ozone long-range transport; for some source-receptor pairs, reducing all three precursors more than doubles the long-range influence of NO_x reductions alone. Relative to reducing NO_x alone, also reducing NMVOCs and CO mainly influences the production of ozone outside of the source region. Reducing NMVOCs and CO causes the global NO_y burden to decrease by reducing the production of PANs, thereby causing long-range influences on ozone. This research suggests that the importance of NMVOCs and CO for long-range transport may have been underemphasized in the past.

There are several important uncertainties and limitations in this work. Our findings are dependent on the definitions of different world regions used here, and should be evaluated through similar experiments in several different models. The emissions used in this study are uncertain and have changed since the early 1990s, particularly in less industrialized regions, and these uncertainties are expected to affect our estimates of the changes in ozone for 10% reductions, particularly for rapidly growing regions such as East Asia, with lesser importance for the effects per ton of emissions. The model is also biased high relative to measured surface ozone concentrations in some NH regions, particularly in summer. Our results are also limited to the 10% anthropogenic emission control experiments simulated here; Wu et al. (2009) find that inter-continental ozone transport in the NH scales nearly linearly with reductions of NO_x in the summer and NMVOCs year-round, but strongly non-linearly with NO_x reductions outside of the summer. Finally, these results are based on one year of typical meteorological conditions, which do not necessarily reflect the multi-year average conditions, nor the conditions for maximum long-range transport. The inter-annual variability in transport conditions should be further studied for multiple source-receptor pairs, beyond the existing studies for particular regions (Duncan and Bey, 2004; Liu et al., 2003, 2005). In addition to improving emission estimates, future research should improve our understanding of the physical

and chemical processes important for the long-range transport of ozone and its precursors, emphasizing the comparison of model results with satellite and in situ measurements, and combining regional and global models.

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