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Effect of regional precursor emission controls on long-range ozone transport – Part 2: steady-state changes in ozone air quality and impacts on human mortality

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Abstract

Large-scale changes in ozone precursor emissions affect ozone directly in the short term, and also affect methane, which in turn causes long-term changes in ozone that affect surface ozone air quality. Here we assess the effects of changes in ozone precursor emissions on the long-term change in surface ozone via methane, as a function of the emission region, by modeling 10% reductions in anthropogenic nitrogen oxide (NO_x) emissions from each of nine world regions. Reductions in NO_x emissions from all world regions increase methane and long-term surface ozone. While this long-term increase is small compared to the intra-regional short-term ozone decrease, it is comparable to or larger than the short-term inter-continental ozone decrease for some source-receptor pairs. The increase in methane and long-term surface ozone per ton of NO_x reduced is greatest in tropical and Southern Hemisphere regions, exceeding that from temperate Northern Hemisphere regions by roughly a factor of ten. We also assess changes in premature ozone-related human mortality associated with regional precursor reductions and long-range transport, showing that for 10% regional NO_x reductions, the strongest inter-regional influence is for emissions from Europe affecting mortalities in Africa. Reductions of NO_x in North America, Europe, the Former Soviet Union, and Australia are shown to reduce more mortalities outside of the source regions than within. Among world regions, NO_x reductions in India cause the greatest number of avoided mortalities per ton, mainly in India itself. Finally, by increasing global methane, NO_x reductions in one hemisphere tend to cause long-term increases in ozone concentration and mortalities in the opposite hemisphere. Reducing emissions of methane, and to a lesser extent carbon monoxide and non-methane volatile organic compounds, alongside NO_x reductions would avoid this disbenefit.

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1 Introduction

Tropospheric ozone has been observed to have increased in polluted regions since preindustrial times, while the global ozone background has also increased (Vingarzan, 2004). Increases in ozone in polluted regions have been attributed mainly to increases in anthropogenic emissions of nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs), and carbon monoxide (CO), while the global background increase has been attributed mainly to anthropogenic NO_x and methane (CH_4) (Wang and Jacob, 1998; Lelieveld and Dentener, 2000). Since NO_x , NMVOCs, and CO react rapidly to form ozone – on the scales of hours to weeks after emissions – the effects of changes in emissions of these precursors on ozone are realized quickly, within a year after the change in emissions. Among the ozone precursors, CH_4 has a significantly longer lifetime – a perturbation lifetime of about 12 years (Forster et al., 2007) – causing ozone concentrations to respond more gradually to sustained changes in CH_4 emissions.

Global reductions in NO_x emissions have been shown previously to cause concentrations of the hydroxyl radical (OH) to decrease (Wang and Jacob, 1998; Fuglestedt et al., 1999; Wild and Prather, 2000; Wild et al., 2001; Fiore et al., 2002). Since OH is the major sink for CH_4 , NO_x reductions cause CH_4 concentrations to increase. For global climate forcing, the increase in CH_4 outweighs the decrease in ozone, causing a net positive radiative forcing from global NO_x emission reductions (Fuglestedt et al., 1999; Wild et al., 2001) and surface NO_x reductions in all world regions (Naik et al., 2005; Berntsen et al., 2005).

The increase in CH_4 concentrations in turn causes a long-term increase in ozone (Wild et al., 2001), on the time scale of the perturbation lifetime of CH_4 . For a sustained decrease in NO_x emissions, this long-term ozone increase will partially counteract the direct short-term ozone decrease. While the long-term increase in ozone has been shown to be important for ozone radiative forcing, it also influences surface air quality; for a 20% decrease in global anthropogenic NO_x emissions, the long-term increase in

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ozone counteracts the short-term ozone decrease by 6–14% for population-weighted ozone air quality metrics (West et al., 2007a). In contrast, reductions in global CO emissions cause a net increase in OH, which causes CH_4 to decrease, leading to a long-term decrease in ozone in addition to the short-term decrease. Global reductions in anthropogenic NMVOC emissions cause nearly negligible effects on CH_4 and the long-term change in ozone, in comparison with short-term ozone changes (West et al., 2007a). Reductions of NO_x from four different world regions have been shown to cause long-term ozone increases that are comparable to the direct short-term ozone decreases due to inter-continental transport, while combinations of NO_x , NMVOC, and CO reductions have much smaller long-term influences due to compensating effects on OH and CH_4 (Fiore et al., 2009).

The companion to this paper (West et al., 2009) evaluates the effects of 10% reductions in anthropogenic NO_x emissions from each of nine world regions on the long-range transport of ozone air pollution, considering only the direct short-term effects on ozone. In this paper, we use those same simulations to evaluate the long-term changes in surface ozone air quality through resulting changes in CH_4 , quantifying the effects of NO_x reductions on CH_4 and long-term ozone for each world region. We then present the sum of the short-term and long-term changes in surface ozone air quality – the net steady-state change for sustained NO_x reductions – and its importance for the ultimate effect on the long-range transport of ozone between all nine world regions. We likewise evaluate the effects of simultaneous reductions in emissions of NO_x , NMVOCs, and CO from three world regions, on CH_4 and long-term surface ozone.

In addition, ground-level ozone has been associated with several health impairments, and with premature human mortality in a large number of epidemiologic daily time series studies (Levy et al., 2001, 2005; Thurston and Ito, 2001; Bell et al., 2004, 2005; Gryparis et al., 2004; HEI International Scientific Oversight Committee, 2004; Ito et al., 2005; Bell and Dominici, 2006; National Research Council, 2008). These effects on human health generally have been the main motivator for ozone air quality regulations. However, the influence of inter-continental ozone transport on human health has not

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been investigated for multiple source-receptor pairs. We therefore evaluate the effects of regional reductions in NO_x emissions on premature human mortality due to ozone, both within each of the nine source regions and due to inter-regional transport for all source-receptor pairs. We first present the methods of estimating the long-term changes in ozone via CH_4 , and the importance of this long-term influence on the net steady-state change in inter-continental ozone transport. Then we present methods of assessing changes in premature human mortality due to changes in ozone, and the avoided mortalities for each world region, for both the short-term change in ozone air quality and the steady-state change.

2 Effects of precursor emission reductions on steady-state ozone air quality

We use the same set of MOZART-2 (Horowitz et al., 2003) simulations that was presented previously by Naik et al. (2005) and the companion to this paper (West et al., 2009), and analyze them here for their effects on long-term changes in surface ozone via changes in atmospheric CH_4 . These simulations were conducted using meteorology from the middle atmosphere version of the Community Climate Model (MACCM3) at 2.8° by 2.8° horizontal resolution, and with emissions for the base simulation that correspond to the early 1990s. Regional emission sensitivity simulations were conducted by removing 10% of ground-level anthropogenic NO_x emissions in each of nine world regions, as defined by Naik et al. (2005) and West et al. (2009), and labeled in Table 1. Three additional simulations considered the simultaneous removal of 10% of anthropogenic NO_x , NMVOCs, and CO in NA, EU, and SE. Emissions have changed since the early 1990s, with likely decreases in anthropogenic ozone precursor emissions from NA, EU, and FSU, and increases elsewhere, particularly in EA (West et al., 2009). This change in emissions will affect our estimates of the change in inter-continental transport and of long-term changes in ozone from 10% reductions, and are likely less important for the changes per unit change in emissions.

Long-term changes in ozone due to changes in CH_4 concentrations are assessed

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following the methods detailed in West et al. (2007a) and Fiore et al. (2009), which are based on Fuglestedt et al. (1999) and Naik et al. (2005). Since the 10% regional changes in anthropogenic NO_x emissions are assumed to be sustained in the future, the long-term change in ozone is added to the direct short-term change to give the net change at steady-state. The change in CH_4 lifetime is assessed from the simulated change in OH; on this basis, we estimate the steady-state change in CH_4 concentration using a CH_4 -OH feedback factor of 1.33, which was diagnosed for MOZART-2 under the same conditions (West et al., 2007a), and is equivalent to the mean of several models (Fiore et al., 2009). We then scale the hourly ozone change in each grid cell from a simulation where anthropogenic CH_4 emissions are reduced by 20% under the same conditions (West et al., 2007a), to the estimated change in CH_4 at steady-state, for each regional NO_x reduction case. We add this long-term change in ozone via CH_4 to the short-term ozone change simulated directly in the model, to give the net change at steady-state; the steady-state ozone is calculated as a full time-varying three-dimensional field, thereby accounting for the spatial distribution on each day of the net steady-state change in ozone. For sustained emission reductions, the net change in ozone will rapidly reflect the short-term change, and gradually approach the steady-state change with a timescale equal to the 12-year perturbation lifetime of CH_4 .

Table 1 shows that the CH_4 increase is greatest for the NO_x reduction from NA, the region with the largest absolute change in NO_x emissions. The greatest CH_4 increase per unit NO_x decrease, however, results in the regions in the tropics and Southern Hemisphere (SE, AU, and SA), with values varying by more than a factor of ten between different regions. These changes in CH_4 are roughly 12% smaller than those reported by Naik et al. (2005) for the same simulations, because we account for CH_4 loss processes other than reaction with OH (losses in the stratosphere and deposition to soil), and we use the CH_4 -OH feedback factor of 1.33 derived from the model rather than the 1.4 recommended by Prather et al. (2001) (see West et al., 2007a).

This greater sensitivity of OH and CH_4 to changes in emissions in tropical regions is in agreement with the finding of a greater change in ozone per ton of NO_x from tropical

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and Southern Hemisphere (SH) emissions, which was attributed to a combination of stronger photochemical activity, greater sensitivity due to lower NO_x concentrations, and more rapid vertical transport that contributes to longer lifetimes for ozone and its precursors (West et al., 2009). These findings also agree with those of Gupta et al. (1998), who show that OH increases as emissions are redistributed toward tropical regions.

The changes in CH₄ drive proportional long-term changes in global surface ozone (Table 1). Again, the 10% NO_x reduction in NA increases long-term surface ozone more than any other region, but the tropical and SH regions have the greatest long-term ozone change per unit change in NO_x. The long-term ozone change will be distributed spatially according to the response of ozone to CH₄, which is fairly uniform globally, but with greater influences in the Northern Hemisphere (NH) than SH, and some regions of peak response, such as the Middle East (West and Fiore, 2005; West et al., 2006, 2007a; Fiore et al., 2008). This long-term global change counteracts the more regionally-focused short-term ozone changes.

Figure 1 shows the effect of including this long-term change in ozone, relative to the short-term change; in many regions, the net steady-state change in ozone is substantially smaller than the short-term change. For the global annual average ozone, the long-term surface ozone change offsets the short-term change by between 13% (for the NO_x reduction in AU) and 89% (for FSU), depending mainly on the magnitude of the short-term change. Here the short-term change in the global annual average ozone is very small for the NO_x reductions from EU and FSU because of local increases in ozone as NO_x is reduced, as discussed by West et al. (2009), particularly in Northern Europe in winter. This local sensitivity causes the global population-weighted ozone to increase for the 10% NO_x reduction in EU, while for FSU it is nearly zero. Figure 1 also shows that the long-term ozone has a stronger influence for the spatially-averaged surface ozone than for the population-weighted, relative to the short-term change, as population-weighted results tend to be more heavily influenced by short-term local changes.

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The net effect of the regional NO_x reductions on ozone long-range transport at steady-state is in Table 2. We show results in the three-month ozone season for each receptor region, as seasonal concentrations are typically of greatest interest for air quality management, and for a population-weighted metric that emphasizes populated regions and is relevant for human health. For all source-receptor pairs, the steady-state influences are less negative (or more positive) than the short term (West et al., 2009). The source-receptor pairs identified as having the greatest long-range transport influence in the short term also have the greatest influence at steady-state (EU on FSU, EA on SE, and EU on AF). While this long-term influence is small in comparison with the short-term effects of a region on itself, it is comparable to the short-term influence for many of the more distant source-receptor pairs. Consequently, while the short-term influences for many distant source receptor pairs indicate a reduction in ozone, the net steady-state influence is near zero or an increase in ozone. For example, the influence of AF on EU is a small decrease in ozone in the short term, but a small increase in ozone at steady-state (Table 2). Likewise, the steady-state influence of EA on NA is roughly half of the short-term influence.

The difference between short-term and steady-state results is illustrated in Fig. 2, focusing on Europe. Figure 2a shows that for both short-term and steady-state results, the 10% NO_x reduction in EU strongly influences FSU, with roughly the same impacts as the influence on EU itself. It also strongly influences AF, while influences on other regions are small. For EU as a source, the differences between short-term and steady-state results are fairly small, between 3 and 5 ppt. Considering EU as a receptor (Fig. 2b), EU is influenced by the 10% NO_x reductions in NA and FSU, with effects that are about 30% as large as the effect of EU on itself. Here, the differences between the short-term and steady-state results are more variable, from 4 to 18 ppt with the reduction in NA causing the greatest difference, as in Table 1.

When these influences on EU are normalized per unit NO_x emissions (Fig. 2c), the effect on EU per ton reduced is greater for NO_x reductions in FSU than in NA, as the FSU has lower total emissions. For both the short-term and steady-state, the benefit

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for EU per ton reduced in FSU is about half of the benefit from reductions in EU itself. Likewise, NO_x reductions in NA are about 20% as effective per ton in reducing ozone in EU, as NO_x reductions from EU itself. In Fig. 2c, the difference between the short-term and steady-state changes per unit NO_x range widely among the different regions, with much larger differences for SE, SA, and AU, in agreement with Table 1. The short-term effect per unit NO_x of SE, SA, and AU on EU is small, but the steady-state influence is larger and negative because of the large influence per ton on CH_4 and long-term ozone. At steady-state, therefore, the dominant influence of NO_x reductions in SE, SA, and AU on EU is to cause ozone to increase, via the long-term increase in CH_4 .

Future studies can estimate the long-term change in ozone due to NO_x reductions from different world regions directly by using the global annual average surface ozone change per unit NO_x emissions in the last column of Table 1, assuming linearity for rather small changes in NO_x . For example, an emission reduction program that would reduce NO_x emissions from NA by 1 TgN yr^{-1} (sustained into the future) would cause a long-term global increase in the annual average surface ozone of about 14 ppt; a reduction in SE would increase ozone by about 96 ppt. Future studies interested in considering the inter-regional influences of the long-term changes in ozone can likewise use Table S2, see <http://www.atmos-chem-phys-discuss.net/9/7079/2009/acpd-9-7079-2009-supplement.pdf>, which includes the spatial distribution of the long-term ozone response to CH_4 . Applications of these results should bear in mind that other models may give different changes in CH_4 and long-term surface ozone (Fiore et al., 2009), and that the responses to large emission changes may not be proportional (Wu et al., 2009).

Table 3 shows the effects of 10% reductions of anthropogenic emissions of NO_x , NMVOCs, and CO on the long-term changes in CH_4 and surface ozone concentration, for three source regions. Relative to only reducing NO_x , reducing three precursors simultaneously creates compensating influences, so the changes in CH_4 are smaller than in Table 1. For the simultaneous reductions from EU, the decrease in CH_4 caused by the NMVOC and CO reductions outweighs the increase from reducing NO_x , for a net

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decrease in CH_4 . In contrast, the simultaneous reductions from NA and SE cause a net increase in CH_4 . This difference arises mainly from the different chemical sensitivities in these regions. If we divide the total CH_4 change attributed to the change in NMVOC and CO emissions by the NMVOC and CO reductions individually (not shown) we find that there is much less variance among these three regions for the CH_4 change due to NMVOCs and CO, than for NO_x . The simultaneous reduction in EU therefore causes CH_4 to decrease mainly because of a lower sensitivity for the NO_x reduction in EU, relative to the other regions (Table 1).

Table 3 then presents the global long-term annual average surface ozone changes resulting from the changes in CH_4 . In Table 4, we show the source-receptor matrix for the net steady-state change in ozone for the simultaneous NO_x , NMVOC and CO reductions. Because the long-term change is very nearly zero for the simultaneous EU reductions (Table 3), there is very little difference from the direct short-term reduction for EU (West et al., 2009). For the simultaneous reductions in NA and SE, the steady-state results are slightly more negative than the short-term results, but these differences between the short-term and steady-state results are much smaller than when only NO_x reductions are considered (Table 2). Similarly, Fiore et al. (2009) found that reducing NO_x , NMVOC and CO emissions in four world regions by 20% causes compensating changes to OH and CH_4 , for little net change in surface ozone; reducing NO_x and CO individually causes more marked long-term ozone changes.

3 Effects of regional emission reductions on premature human mortality

Here we assess the effects of inter-continental ozone transport on the incidence of premature human mortality, using the relationships between ozone concentration and mortality from epidemiologic daily time-series studies. We employ the methods used previously by West et al. (2006, 2007b) to assess the global effects of ozone concentration changes on mortality, consistent with methods recommended by the National Research Council (2008). We use the the ozone-mortality relationship derived from

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a daily time-series study that uses a large database of 95 cities in the United States (US) (Bell et al., 2004). This ozone-mortality relationship (given by β) is smaller than reported by recent meta-analyses (Bell et al., 2005; Ito et al., 2005; Levy et al., 2005), and is chosen for its large dataset with no possibility of publication bias. While the relationship between ozone and mortality has not been investigated thoroughly in developing nations, similar relationships have been found in other studies in the US (Levy et al., 2001; Thurston and Ito, 2001; Bell and Dominici, 2006), Europe (Gryparis et al., 2004), and some of the few available studies in developing nations (Borja-Aburto, 1998; Kim et al., 2004; O'Neill et al., 2004; HEI International Scientific Oversight Committee, 2004).

We estimate the change in total non-accidental mortality in each grid cell on each day, using the change in 24-hour average ozone concentration (for both the short-term and steady-state ozone changes), using a value for β of 0.52% excess non-accidental mortalities per 10 ppb change in 24-hour ozone (Bell et al., 2004). Available epidemiologic studies do not clearly show a low-concentration threshold below which changes in ozone have no effect on mortality (Bell and Dominici, 2006; National Research Council, 2008); here we assume a low-concentration threshold for ozone of 25 ppb, as in previous studies (West et al., 2006, 2007b).

The global distribution of population in 2003 is taken from the LandScan database (Oak Ridge National Laboratory, 2005), and mapped onto the MOZART-2 modeling grid. Non-accidental baseline mortality rates are taken from the World Health Organization (2004) for each of 14 world regions, which are also mapped onto the modeling grid. Estimates of avoided mortalities have large uncertainty due to the uncertainties in β , the low-concentration threshold, and the application of the ozone-mortality relationship in less industrialized nations, in addition to the uncertainty in the modeled ozone response itself, and these uncertainties have been explored elsewhere West et al. (2006, 2007b). Here we emphasize the relative changes in mortality in different receptor regions, due to regional emission reductions and inter-regional transport, rather than the total numbers of estimated mortalities. We test the sensitivity of our major

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findings for mortalities due to only cardiovascular and respiratory (CR) mortalities, and for alternative low-concentration thresholds.

3.1 Results

We first show results for direct short-term changes in ozone, as these would be realized immediately, while the steady-state changes in ozone would be experienced by a future population. Table 5 shows the effects of regional NO_x reductions on the avoided premature all-cause mortalities per year resulting from short-term changes in ozone in each region, as a source-receptor matrix. The avoided mortalities estimated in each region are influenced by the population, the baseline mortality rate, and the daily changes in ozone in each grid cell, for days above the threshold. The results highlight the receptor regions EA, IN, and AF, as these regions have the highest population, and AF has the highest baseline mortality rate in the world. The results show that the 10% reduction in NO_x emissions in IN causes the greatest number of avoided mortalities, with 93% of that benefit occurring in IN itself. Large numbers of avoided mortalities result in IN from NO_x reductions within the same region because of the high population density and because it has the largest decrease in the population-weighted annual average ozone (Fig. 1b); NO_x reductions in IN also reduce ozone throughout the year, while reductions in NA, EU, FSU, and EA increase the population-weighted ozone in the NH winter (West et al., 2009).

Decreases in NO_x from many regions cause substantial reductions in mortalities in other regions. In particular, the 10% NO_x reductions in NA, AF, EA, and SE all have substantial influences on mortality in IN. The greatest inter-regional influence on mortality is from the 10% reduction in EU on mortalities in AF, where most of those avoided mortalities are in the Middle East and North Africa.

Interestingly, the NO_x reduction in NA causes a substantial number of avoided premature mortalities in the regions AF, EU, IN, and EA, such that more avoided premature mortalities result outside of NA than within. Similarly, the NO_x reductions in EU, FSU, and AU also cause a greater number of avoided premature mortalities outside of these

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source regions than within, consistent with other results for Europe using a different atmospheric model (Duncan et al., 2008). This results in part because the populations influenced elsewhere in the NH are much greater than within the NA, EU and FSU source regions. Similarly, AU has a small population within that region, but larger populations potentially exposed in AF, SE, and SA. Further, the regions NA, EU, and FSU all contain areas where NO_x reductions cause increases in ozone, particularly in populated areas in winter, which decreases the number of intra-regional avoided mortalities. While some winter days may be below the low-concentration threshold, therefore having no effect on the estimated avoided mortalities, many such days are above this threshold. The reduction in EU is actually estimated to increase mortalities within that same region, as the population-weighted annual average concentration increases (Fig. 1b) due to an increase in ozone in Northern Europe and a decrease near the Mediterranean Sea. Summing over the 10% reductions in all nine regions, we find that the total inter-regional avoided mortalities are substantial: 2600 per year, or 31% of the total avoided mortalities.

In Table 6, we normalize the avoided premature mortalities per unit NO_x emissions reduced in each source region. As in Table S1, estimating the reduction per unit NO_x emissions emphasizes source regions in the tropics and SH, due to the greater sensitivity to NO_x reductions. The results show that an average ton of NO_x reduced in IN results in the greatest reduction in mortalities, where the great majority of these avoided mortalities are within IN itself. Reducing emissions in SE also has a large effect on the avoided premature mortalities per unit NO_x, with large impacts on avoided mortalities in the heavily populated regions IN and EA.

Table 7 shows that the simultaneous reduction of NO_x, NMVOCs and CO causes many more avoided premature mortalities than when reducing NO_x alone. The conclusion that NO_x reductions in NA and EU cause more avoided mortalities outside of these regions than within also holds when reducing all three precursors. These results are compared with those of Casper-Anenberg et al. (2009) who assess mortality for simulations from the HTAP multimodel intercomparison (Fiore et al., 2009), where we

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use the same conditions and definitions of receptor regions as in that study (Table S3). Results show fewer intra-regional and inter-regional avoided mortalities in the present study, which is attributed mainly to the lower sensitivity of MOZART-2 to changes in precursor emissions than the HTAP multimodel mean (West et al., 2009), as the two studies use very similar methods to assess mortalities for a given change in ozone concentration.

Figure 3 and Tables 8, S4, and S5 show results for avoided mortalities at steady-state. We calculate the steady-state avoided mortalities using the present-day population, for ease of comparison with the short-term results; since population is expected to grow, the ultimate inter-continental effects on mortality may be greater than represented here, particularly for tropical receptor regions. Figure 3 shows that at steady-state, avoided mortalities are widespread in the NH for the 10% NO_x reductions from NA, EU, FSU, IN, and EA. These reductions also result in small increases in mortality in the SH, due to the long-term ozone increase. Similarly reductions from SA, SE, and AU decrease mortality in the SH, but increase mortality in much of the NH, as the long-term ozone increase dominates in the NH.

Comparing Table 8 at steady-state with the short-term results (Table 5), we see that the results are similar, but with fewer avoided mortalities. Because the long-term ozone increase is globally widespread, the largest differences between the short-term and steady-state results are in the inter-regional mortalities, which decrease substantially from 2600 per year in Table 5 to 1432 here (20% of the total), accounting for most of the difference in total mortalities between the two cases. Because of the large effect on long-term ozone from emissions in the tropics and SH, we see that the NO_x reduction from AU causes a net increase in mortalities at steady-state, mainly because of mortality increases in highly populated regions of the NH. Figure 3 also shows localized increases in mortalities within the source regions of NA, EU, FSU, and EA, which are generally caused by local increases in ozone, as discussed earlier, and are often strongest in winter.

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3.2 Sensitivity

While the overall uncertainty in estimates of mortality is substantial, and influenced by several assumptions and parameters (West et al., 2007b), we are most interested here in the uncertainties and assumptions important for the relative numbers of avoided mortalities among the source-receptor pairs. In particular, we analyze the conclusion that 10% reductions in anthropogenic NO_x emissions in NA, EU, and FSU cause more avoided premature mortalities outside of these source regions than within.

An important assumption for the relative numbers of mortalities within and outside of the source region is that the concentration-response relationship is applicable globally. As discussed earlier, the concentration-response relationship is estimated in one large study in the US, which is broadly consistent with other studies in North America and Europe. Fewer studies are available in less industrialized nations for comparison, but the large difference in general causes of death in less industrialized nations suggests that these concentration-response relationships may be inaccurate elsewhere.

As a sensitivity analysis, we consider only the effects of ozone on CR mortality, since CR causes are the most plausible means by which ozone might affect mortality. In less industrialized nations, baseline CR mortality rates are generally much lower than the baseline non-accidental mortality rate, since a large fraction of the population dies from other causes. We estimate CR mortality using the appropriate CR baseline mortality rate for each of the 14 world regions (World Health Organization, 2004), and the β for CR mortality from Bell et al. (2004). In Table 9, the global avoided CR mortalities decrease relative to the total mortality base case, for the 10% NO_x reductions from NA, EU, and FSU, as a result of the lower baseline CR mortality rates. Here we show mortality estimates at steady-state, since the steady-state results have greater within-region avoided mortalities as a fraction of the total (Table 8). For all three regions, more avoided CR mortalities occur outside of the source region than within.

Second, we consider the uncertainty in the low-concentration threshold, below which changes in ozone are assumed to not affect mortality. Reductions in NO_x from both NA

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and EU cause some local increases in ozone near areas with large concentrations of population and emissions (Fig. 3). Since many of these increases occur on days with low ozone concentrations and in winter, raising the threshold would cause a smaller increase in mortalities in these areas. By raising the threshold from 25 ppb to 40 ppb, the avoided mortalities within the source regions increase for all three regions in Table 9. For EU and FSU, the conclusion holds that there is a greater reduction in mortality outside of the source region than within. For NA, however, the avoided mortalities within the source region exceed those outside of the source region, when using the 40 ppb threshold.

Similarly, rather than increasing the threshold, we exclude the winter months, considering only seven months from April through October. Some analyses of ozone mortality in NH temperate regions consider only the summer months when ozone pollution is highest. However, these same seasonal patterns for ozone do not hold globally, particularly in tropical regions. By excluding the winter months, within-region mortalities increase relative to the base case (Table 9), and the conclusion that most avoided mortalities are outside of EU and FSU still holds. For NA, the NO_x reduction causes more avoided mortalities within (versus outside of) the source region.

4 Conclusions

Large-scale changes in emissions of ozone precursors cause direct short-term changes in ozone that affect air quality on inter-continental scales. These changes also cause global long-term changes in ozone via changes in CH_4 . Here we quantify these long-term effects on surface ozone air quality as a function of the source region, for nine source regions globally. Reductions in NO_x emissions from all source regions are shown to increase CH_4 and ozone in the long term. While the long-term increase in ozone is small compared with the short-term ozone decrease within a source region, it is comparable to many of the short-term inter-regional influences, but opposite in sign. For some source-receptor pairs, the long-term increase is roughly equal to or exceeds

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the short-term decrease, for a net increase in ozone. For example, the steady-state influence of NO_x reductions in EA on NA is roughly half of the short-term influence. Decreases in NMVOC and CO emissions reduce CH₄ and cause a decrease in ozone in the long term. Per unit decrease in NO_x emissions, the long-term increase in ozone is greatest from source regions in the tropics and SH, in some cases more than a factor of ten greater than the long-term increase from reductions in temperate regions of the NH. The dominant influence of NO_x reductions in tropical and SH source regions on temperate regions of the NH is the long-term ozone increase. Our estimates of the long-term ozone increase can be used in future studies to add to the short-term effects of ozone precursor reductions modeled directly on regional scales.

We also estimate the avoided premature mortalities in all world regions due to changes in ozone from regional reductions of precursor emissions. The strongest inter-regional effect on mortality is seen for EU on AF, where most of these avoided mortalities are in the Middle East and North Africa; this is true for short-term and steady-state cases, as well as whether emissions of NO_x or all three precursors are reduced. For 10% NO_x reductions in all regions, we find that 31% of the total avoided mortalities in the short term are due to inter-regional transport; although the change in ozone within the source region is generally much greater than elsewhere, a much larger population is potentially exposed outside of the source region. The contribution of inter-regional transport decreases to 20% of total mortalities when the long-term increase in ozone (via CH₄) is accounted for. Reducing NO_x emissions from IN is shown to cause the most avoided premature mortalities per ton of NO_x of any world region, with most of those avoided mortalities occurring within IN itself, due to the high population density and strong sensitivity to changes in NO_x.

Reductions in ozone precursor emissions in EU, NA, and FSU are shown to cause more ozone-related avoided premature mortalities outside of the source region than within, due to both larger relevant populations outside of the source region, and because NO_x reductions cause ozone to increase in some locations within the source region, mainly in winter. These findings agree with analyses for EU using a different

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atmospheric model (Duncan et al., 2008), and for EU and NA using a multimodel ensemble mean (Casper-Anenberg et al., 2009). This analysis is limited to considering ozone-related mortality; including changes in particulate matter (PM) concentrations due to these precursor reductions may significantly change the results, as PM has been strongly linked with mortality (Pope et al., 2002).

This conclusion that reductions in EU, NA, and FSU cause more avoided mortalities outside of these regions than within is robust over sensitivities to key parameters and assumptions in our mortality estimates, except for NA in some cases considered. It is also subject to uncertainties in estimates of the relative changes in ozone within and outside of the source region, from the atmospheric model. In their multimodel intercomparison, (Fiore et al., 2009) find that there is rather little disagreement among different models in the effects of inter-continental transport, but much greater disagreement in impacts of emission reductions within the source regions, as global models with different emissions have different local sensitivities. Casper-Anenberg et al. (2009) show that for this multimodel ensemble, the relative mortalities within versus outside of source region is strongly influenced by uncertainty in the intra-regional change in ozone among the different models. While a global model is appropriate for addressing long-range transport, estimates of the population-weighted change in concentration within a source region (and mortalities) should be analyzed by regional models at finer scales, that have been tested extensively for local conditions. Our assessment of the effects of long-range transport on mortality is also limited by our understanding of the mortality effects of ozone, particularly in developing nations, and future research should aim to improve this understanding.

We show that accounting for the long-term influence of precursors on ozone can change estimates of the net effect on mortality, and may be the dominant influence for more distant source-receptor pairs. For example, reductions of NO_x in the SH are expected to generally increase mortalities at steady-state in the NH. Because of the long-term global ozone increase resulting from NO_x reductions, as well as the net increase in radiative climate forcing, ozone air quality management may consider a greater em-

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phasis on reducing NMVOCs, CO, and particularly CH₄, alongside reductions in NO_x (Fiore et al., 2002; West et al., 2007a). Finally, these findings of large health benefits outside of source regions suggest that national or regional assessments of the costs and benefits of air pollutant control actions (such as are conducted routinely by the US Environmental Protection Agency or the European Commission) may significantly underestimate the total human health benefits by not including benefits outside of national or continental boundaries.

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Table 1. Long-term changes in global annual mean CH₄ and surface O₃ concentrations, due to 10% regional reductions in anthropogenic NO_x emissions.

Source Region	Change in global CH ₄ (ppb)	CH ₄ change per Tg decrease in NO _x emissions (ppb (TgN yr ⁻¹) ⁻¹)	Change in surface O ₃ (ppt)	Surface O ₃ change per Tg decrease in NO _x emissions (ppt (TgN yr ⁻¹) ⁻¹)
NA – North America	4.54	5.7	11.5	14
EU – Europe	1.30	2.7	3.3	7
FSU – Former Soviet Union	0.99	4.0	2.5	10
AF – Africa	2.73	13.7	7.0	35
IN – India	1.55	13.5	3.9	34
EA – East Asia	2.31	5.8	6.0	15
SA – South America	2.51	30.6	6.5	79
SE – Southeast Asia	2.49	38.0	6.3	96
AU – Australia	1.21	31.7	3.1	81

7102

Table 2. Source-receptor matrix of the steady-state change in the 3-month population-weighted average ozone concentration (ppt), for the 10% regional NO_x emission reductions. Results are shown for the consecutive three-month period with highest population-weighted ozone for each receptor region, as defined by West et al. (2009). The diagonal, showing the effect of each region on itself, is underlined, while the most negative values off of the diagonal are bold.

		Receptor Region								
		NA	EU	FSU	AF	IN	EA	SA	SE	AU
Source Region	NA	<u>-498</u>	-45	-33	-30	-29	3	1	2	9
	EU	-4	<u>-189</u>	-180	-69	-9	-6	4	1	4
	FSU	-11	-51	<u>-398</u>	-13	-13	-25	3	2	3
	AF	4	2	-7	<u>-167</u>	-41	6	-1	-1	-10
	IN	1	6	2	-1	<u>-476</u>	-13	2	-28	2
	EA	-9	2	-10	1	1	<u>-924</u>	5	-99	5
	SA	2	11	8	4	4	7	<u>-245</u>	2	-28
	SE	8	12	8	6	-7	-32	-2	<u>-259</u>	-4
	AU	4	5	4	3	4	3	-10	0	<u>-176</u>

7103

Table 3. Long-term changes in global annual mean CH₄ and surface O₃ concentrations, due to 10% regional reductions in anthropogenic emissions of NO_x, NMVOCs, and CO combined.

Source Region	Change in global CH ₄ (ppb)	CH ₄ change from 10% NMVOC and CO reductions (ppb) ^a	Change in surface O ₃ (ppt)	Surface O ₃ change from 10% NMVOC and CO reductions (ppt) ^a
NA	2.30	-2.23	5.8	-5.7
EU	-0.13	-1.44	-0.4	-3.7
SE	1.81	-0.68	4.7	-1.6

^a Inferred as the difference between the 10% reduction of NO_x, NMVOCs, and CO, and only reducing NO_x (Table 1).

7104

Table 4. Source-receptor matrix of the steady-state change in the 3-month population-weighted average ozone concentration (ppt), for the 10% regional emission reductions of NO_x, NMVOCs, and CO.

		Receptor Region								
		NA	EU	FSU	AF	IN	EA	SA	SE	AU
Source Region	NA	<u>-660</u>	-104	-70	-78	-72	-12	-10	-23	-1
	EU	<u>-30</u>	<u>-407</u>	-240	-134	-40	-18	-2	-21	-2
	SE	2	<u>6</u>	4	-2	-19	-43	-10	<u>-296</u>	-12

7105

Table 5. Avoided premature non-accidental mortalities per year due to reductions in 10% of anthropogenic NO_x emissions from each region, for short-term changes in ozone.

		Receptor Region									
		NA	EU	FSU	AF	IN	EA	SA	SE	AU	Global
Source Region	NA	<u>251</u>	148	59	162	133	106	11	7	0	876
	EU	12	<u>-289</u>	89	250	39	54	0	2	0	158
	FSU	12	<u>53</u>	<u>50</u>	67	62	89	0	1	0	333
	AF	12	49	<u>36</u>	<u>938</u>	134	58	4	5	5	1238
	IN	13	13	10	<u>53</u>	<u>3012</u>	80	1	56	0	3238
	EA	38	45	25	34	107	<u>1154</u>	0	124	0	1527
	SA	3	-1	0	33	9	<u>-1</u>	<u>203</u>	3	1	251
	SE	3	1	1	29	149	100	<u>4</u>	<u>417</u>	0	704
	AU	-1	-1	0	7	-2	-2	5	<u>7</u>	<u>7</u>	20
	Global ^a										<u>8344</u>

^a Global is the sum of 10% reductions in each region, and may differ from the results of a simulation where anthropogenic NO_x is reduced by 10% globally.

7106

Table 6. Avoided premature mortalities (Table 5) per year per Tg reduction in NO_x emissions (mortalities (TgNyr⁻¹)⁻¹), for short-term changes in ozone.

		Receptor Region									
		NA	EU	FSU	AF	IN	EA	SA	SE	AU	Global
Source Region	NA	<u>32</u>	18	7	20	17	13	1	1	0	110
	EU	3	<u>-60</u>	18	52	8	11	0	4	0	33
	FSU	5	<u>21</u>	<u>20</u>	27	25	36	0	0	0	135
	AF	6	25	<u>18</u>	<u>472</u>	68	29	2	0	0	623
	IN	12	11	8	46	<u>2621</u>	70	1	3	0	2818
	EA	10	11	6	8	<u>27</u>	<u>290</u>	0	49	0	383
	SA	4	-1	0	40	12	<u>-1</u>	<u>248</u>	31	1	306
	SE	5	2	1	44	227	152	<u>6</u>	<u>637</u>	0	1076
	AU	-1	-2	-1	18	-6	-4	13	<u>17</u>	<u>19</u>	52
	Global ^a										345

^a Here, global is the average avoided mortalities per unit NO_x reduced globally, taken as the sum of avoided mortalities in all nine regions, divided by 10% of the global anthropogenic NO_x emissions.

7107

Table 7. Avoided premature mortalities per year due to reductions in 10% of anthropogenic NO_x, NMVOCs, and CO emissions from each region, for short-term changes in ozone.

		Receptor Region									
		NA	EU	FSU	AF	IN	EA	SA	SE	AU	Global
Source Region	NA	<u>501</u>	259	109	318	246	247	15	22	0	1719
	EU	<u>52</u>	<u>83</u>	179	492	124	217	2	15	0	1164
	SE	10	<u>9</u>	5	64	206	136	7	<u>467</u>	0	905

7108

Table 8. As Table 5, but at steady-state.

		Receptor Region									
		NA	EU	FSU	AF	IN	EA	SA	SE	AU	Global
Source Region	NA	<u>231</u>	117	44	73	59	49	4	-6	0	571
	EU	7	<u>-298</u>	84	225	18	37	-2	-2	0	70
	FSU	7	46	<u>47</u>	48	46	77	-2	-2	0	267
	AF	0	31	27	<u>885</u>	90	24	0	-3	0	1055
	IN	7	2	5	23	<u>2987</u>	61	-42	52	0	3134
	EA	28	29	17	-11	69	<u>1125</u>	-3	118	0	1371
	SA	-8	-18	-8	-16	-32	<u>-32</u>	199	-4	1	82
	SE	-8	-15	-7	-20	108	69	0	<u>410</u>	0	537
	AU	-6	-9	-4	-17	-22	-17	3	<u>3</u>	<u>7</u>	-62
	Global										

7109

Table 9. Sensitivity analysis of avoided premature mortalities globally and within the source region (in parenthesis) from 10% regional NO_x reductions at steady-state, in the base case (from Table 8), and considering cardiovascular and respiratory mortality (CR) only, a low-concentration threshold of 40 ppb, and excluding winter (for April–October only).

		Base case	CR only	Threshold 40 ppb	Excluding winter
Source Region	NA	571 (231)	360 (130)	534 (347)	468 (299)
	EU	70 (-298)	38 (-190)	609 (281)	380 (-14)
	FSU	267 (47)	193 (41)	271 (95)	426 (167)

7110

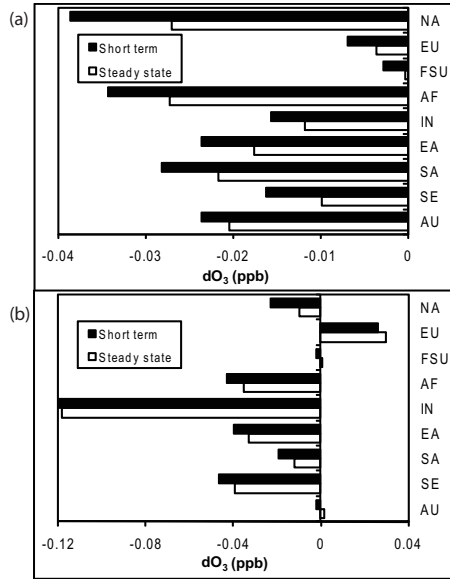


Fig. 1. Change in surface ozone due to 10% reductions of NO_x emissions in each of nine regions, for the short term and at steady-state, for **(a)** global annual average ozone, and **(b)** global population-weighted annual average ozone.

7111

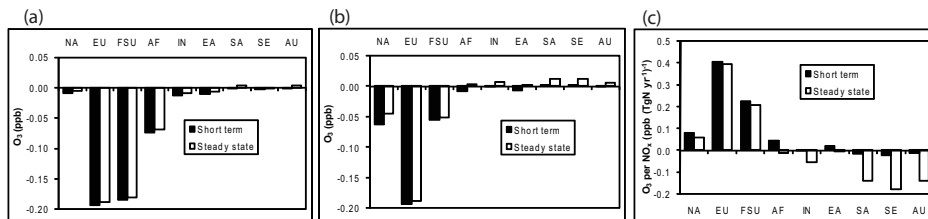


Fig. 2. EU as a source and receptor, using the change in 3-month (JJA) population-weighted O_3 in each receptor region, for the short term and steady-state (from West et al. (2009) for short-term results, and Tables 2 and S1 for steady-state results): **(a)** EU as a source affecting nine receptor regions (reading across Table 2), **(b)** EU as a receptor affected by nine source regions (reading down Table 2), and **(c)** EU as a receptor per unit change in NO_x (reading down Table S1).

7112

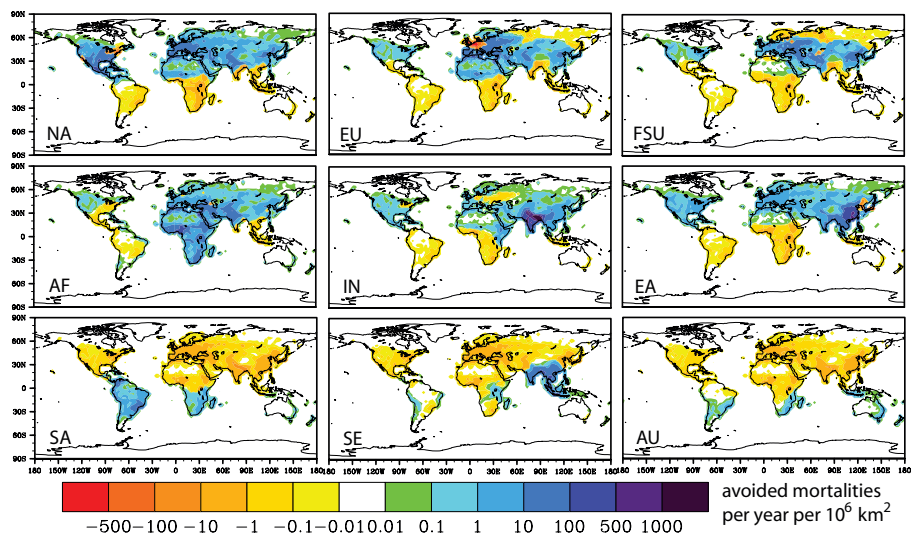


Fig. 3. Avoided premature mortalities from 10% anthropogenic NO_x reductions in each of nine world regions, at steady-state and using a low-concentration threshold of 25 ppb.