Ozone air quality and radiative forcing consequences of changes in ozone precursor emissions

J. Jason West,[1,2,3] Arlene M. Fiore,[4] Vaishali Naik,[2] Larry W. Horowitz,[4] M. Daniel Schwarzkopf,[4] and Denise L. Mauzerall[2,5]

Received 21 December 2006; revised 8 February 2007; accepted 22 February 2007; published 27 March 2007.

[1] Changes in emissions of ozone (O_3) precursors affect both air quality and climate. We first examine the sensitivity of surface O_3 concentrations (O_3^{srf}) and net radiative forcing of climate (RF_{net}) to reductions in emissions of four precursors – nitrogen oxides (NO_x), non-methane volatile organic compounds, carbon monoxide, and methane (CH_4). We show that long-term CH_4-induced changes in O_3, known to be important for climate, are also relevant for air quality; for example, NO_x reductions increase CH_4, causing a long-term O_3 increase that partially counteracts the direct O_3 decrease. Second, we assess the radiative forcing resulting from actions to improve O_3 air quality by calculating the ratio of ΔRF_{net} to changes in metrics of O_3^{srf}. Decreases in CH_4 emissions cause the greatest RF_{net} decrease per unit reduction in O_3^{srf}, while NO_x reductions increase RF_{net}. Of the available means to improve O_3 air quality, therefore, CH_4 abatement best reduces climate forcing. **Citation:** West, J. J., A. M. Fiore, V. Naik, L. W. Horowitz, M. D. Schwarzkopf, and D. L. Mauzerall (2007), Ozone air quality and radiative forcing consequences of changes in ozone precursor emissions, *Geophys. Res. Lett.*, *34*, L06806, doi:10.1029/2006GL029173.

1. Introduction

[2] Concentrations of ozone (O_3) have increased historically at the surface in industrialized regions, and in the global background troposphere [*Vingarzan*, 2004]. Actions to decrease tropospheric O_3 to improve air quality have emphasized reducing emissions of short-lived O_3 precursors – nitrogen oxides (NO_x), non-methane volatile organic compounds (NMVOCs), and carbon monoxide (CO) – on urban and regional scales. Methane (CH_4), a long-lived O_3 precursor (lifetime ~9 yr), contributes to the global background concentration of tropospheric O_3, and CH_4 emission controls have received recent attention as a means of international O_3 air quality management [*Fiore et al.*, 2002; *Dentener et al.*, 2005; *West and Fiore*, 2005; *West et al.*, 2006].

[3] Reducing emissions of any precursor to improve O_3 air quality also influences climate, as such changes affect the concentrations of and radiative forcing from both O_3 and CH_4. Reductions in surface NO_x emissions, for example, decrease O_3 and global concentrations of the hydroxyl radical (OH). Since reaction with OH is the major sink for CH_4, NO_x reductions increase the atmospheric lifetime and concentrations of CH_4. The resulting positive radiative forcing from increased CH_4 roughly cancels, or slightly exceeds, the negative forcing from decreased O_3 globally [*Fuglestvedt et al.*, 1999; *Wild et al.*, 2001; *Fiore et al.*, 2002; *Berntsen et al.*, 2005; *Naik et al.*, 2005; *Shindell et al.*, 2005]. In contrast, reductions in emissions of NMVOCs, CO, and CH_4 increase OH and decrease the forcing from both O_3 and CH_4 [*Prather et al.*, 2001; *Fiore et al.*, 2002].

[4] These changes in CH_4 will also affect O_3 on the longer time scale of the CH_4 perturbation lifetime. In the case of NO_x controls, the CH_4 increase induces a long-term O_3 increase, which partially counteracts the short-term O_3 decrease. Conversely, controls on NMVOCs and CO cause a long-term O_3 decrease in addition to the short-term O_3 decrease. These long-term changes in O_3 have been included in estimates of net radiative forcing [*Wild et al.*, 2001; *Berntsen et al.*, 2005; *Naik et al.*, 2005]; here we also quantify their relevance for long-term surface O_3 air quality.

[5] Local actions to improve O_3 air quality therefore affect surface O_3 and radiative forcing on global scales; while the global effects of emission changes in a single airshed are expected to be small, the cumulative effects of such actions in many polluted regions are analyzed here. The first objective of this study is to simulate the sensitivity of surface O_3 concentrations, and of the net radiative forcing due to O_3 and CH_4, to global emission reductions of NO_x, NMVOCs, CO, and CH_4, using consistent methods. In doing so, we account for the long-term changes in O_3 via CH_4. These estimates can be used together with information on the costs and feasibility of reducing precursor emissions, in the integrated planning of actions to address O_3 air quality and climate change. Changes in aerosol radiative forcing are not included in our estimate of net radiative forcing, but are estimated here to be small (section 3.1.). The second objective addresses the effects of actions to improve surface O_3 air quality on radiative forcing. Because precursor emissions are being reduced in many nations to improve O_3 air quality, and because such actions affect climate, we compare the radiative forcing consequences of decreasing surface O_3 through reductions in emissions of each precursor. We normalize the change in net radiative forcing by changes in surface O_3 concentration metrics, and

[1] Atmospheric and Oceanic Sciences Program, Princeton University, Princeton, New Jersey, USA.
[2] Woodrow Wilson School of Public and International Affairs, Princeton University, Princeton, New Jersey, USA.
[3] University of North Carolina at Chapel Hill, Chapel Hill, North Carolina, USA.
[4] NOAA Geophysical Fluid Dynamics Laboratory, Princeton, New Jersey, USA.
[5] Department of Geosciences, Princeton University, Princeton, New Jersey, USA.

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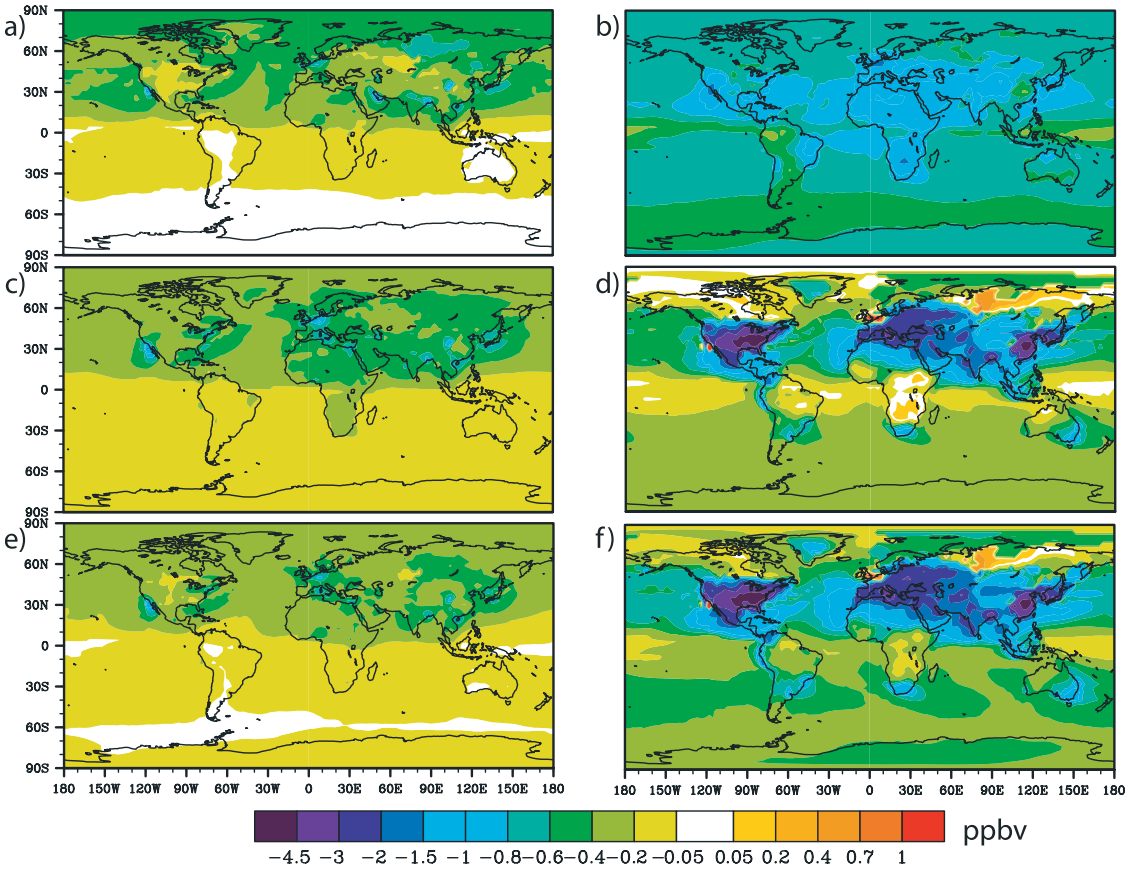

Figure 1. The change in 8-hr. daily maximum surface O_3 concentrations, averaged over the "O_3 season" (the three-month period with highest O_3 in each grid cell), due to 20% reductions in global anthropogenic emissions of O_3 precursors. Results are shown at steady state for (a) NMVOCs, (b) CH_4, (c) CO, and (d) NO_x, and short-term responses are shown for (e) CO and (f) NO_x. The short-term response for NMVOCs is nearly identical to the steady-state response.

present this ratio as an indicator of the climate forcing resulting per unit improvement in O_3 air quality.

2. Methods

[6] We use the MOZART-2 global three-dimensional model of tropospheric chemistry and transport, driven with meteorology from the middle atmosphere version of the Community Climate Model (MACCM3), with a horizontal resolution of about 2.8° by 2.8° and 34 vertical levels. Emissions in the base simulation are representative of the early 1990s. The base simulation includes only minor model updates from previous model versions [*Horowitz et al.*, 2003; *Naik et al.*, 2005] that have been thoroughly evaluated with measurements [*Horowitz et al.*, 2003].

[7] We simulate the effects of sustained uniform 20% reductions in global anthropogenic emissions of four precursors individually (NO_x, NMVOCs, CO, and CH_4), relative to the base simulation. The anthropogenic sources reduced in these experiments include the combustion of biofuels (among other sources), but exclude other forms of biomass burning. Changes in emissions of each gas-phase precursor are assumed to occur independently; we neglect possible changes in emissions of co-emitted species due to the control action. A fixed global CH_4 mixing ratio of 1700 ppbv is used in the base and NO_x, NMVOC, and CO control simulations. In the CH_4 control case, we decrease the CH_4 mixing ratio to 1460 ppbv, the steady-state mixing ratio that would result from a 20% anthropogenic emission reduction, based on the CH_4-OH feedback factor of 1.33 derived from the model (see auxiliary material).[1] All simulations are conducted for 25 months, with results presented for the final 12 months.

[8] The short-term O_3 response to changes in NO_x, NMVOC, and CO is simulated directly in the model, and is assumed constant into the future. This short-term O_3 change adds to the long-term change via CH_4 (the "primary mode" [*Prather*, 1996]) to give the net O_3 change at steady state [*Wild et al.*, 2001]. Previous studies have typically focused only on the short-term O_3 effects of NO_x, NMVOC and CO reductions [*Fuglestvedt et al.*, 1999; *Fiore et al.*, 2002], or used other model results to estimate the long-term response [*Berntsen et al.*, 2005; *Naik et al.*, 2005]. We estimate the long-term O_3 response by scaling the hourly O_3 change in each model grid cell from our CH_4 emission reduction simulation to the implied CH_4 changes derived from the other precursor reduction simulations (based on the simulated change in CH_4 lifetime and the CH_4-OH feedback factor; see equations in auxiliary material). Adding the estimated long-term change in O_3 to the simulated short-

[1] Auxiliary material data sets are available at ftp://ftp.agu.org/apend/gl/2006gl029173. Other auxiliary material files are in the HTML.

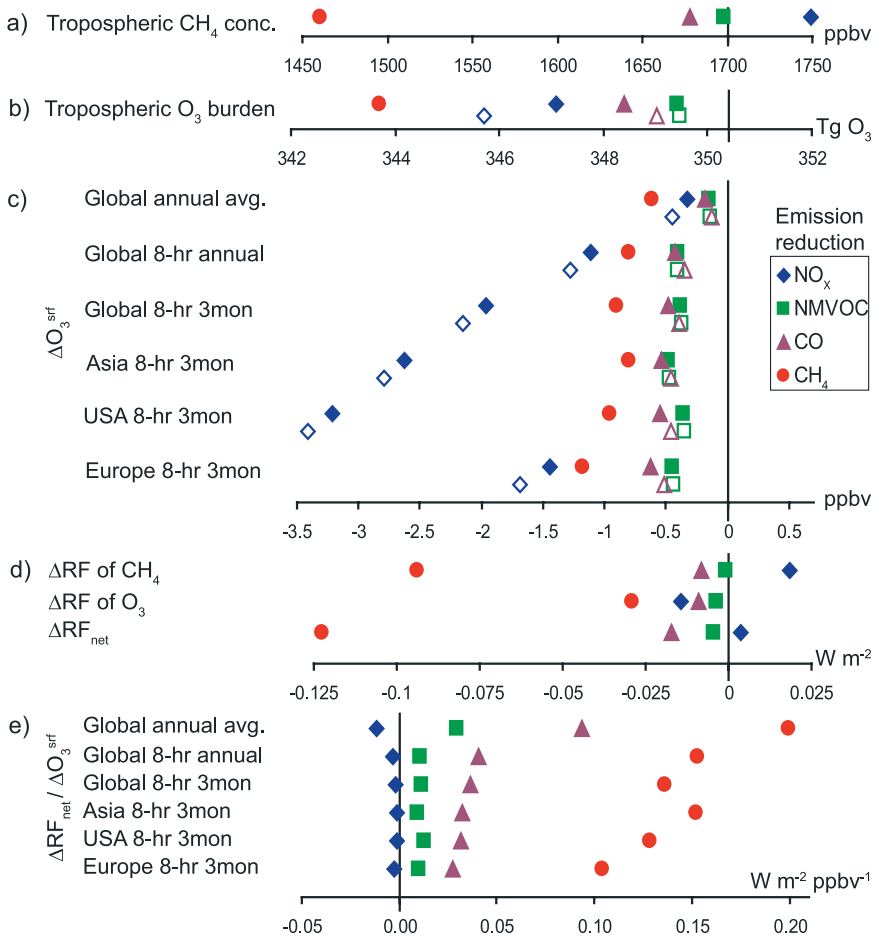

Figure 2. Effects of sustained 20% reductions in global anthropogenic emissions of four O_3 precursors on metrics of (a) CH_4 concentration, (b) O_3 burden, (c) surface O_3 concentration, (d) radiative forcing (RF), and (e) $\Delta RF_{net}/\Delta O_3^{srf}$. Steady-state results are shown with solid symbols; open symbols indicate short-term results (the CH_4 perturbation only causes a steady-state change). Base simulation values are shown by the vertical lines (see auxiliary material). Air quality ΔO_3^{srf} metrics include the population-weighted 8-hr. O_3, annually averaged globally and averaged over the three-month O_3 season globally and regionally (all 8-hr. metrics shown are population-weighted). Radiative forcing indicates the global annual mean forcing at the tropopause.

term change, and assuming O_3 scales linearly with CH_4, is supported by previous model results [*Fiore et al.*, 2002; *Shindell et al.*, 2005].

[9] Long-term O_3 changes will gradually approach their final response according to the modeled perturbation lifetime of CH_4 of 12.1 yr, reaching 56% of the final response in 10 yr, 81% in 20 yr, and 92% in 30 yr. Projected changes in emissions of O_3 precursors over 30 yr are expected to have only minor effects on the CH_4-O_3 sensitivity [*West et al.*, 2006] and on the CH_4-OH feedback factor [*Prather et al.*, 2001].

[10] The global annual mean radiative forcing is calculated as the instantaneous forcing (long-wave plus short-wave) at the tropopause. We calculate the radiative forcing from O_3 using monthly mean O_3 fields at steady state and the Geophysical Fluid Dynamics Laboratory radiative transfer model [*Freidenreich and Ramaswamy*, 1999; *Schwarzkopf and Ramaswamy*, 1999; *Geophysical Fluid Dynamics Laboratory Global Atmospheric Model Development Team*, 2004], as employed previously by *Naik et al.* [2005]. The radiative forcing from CH_4 perturbations is estimated at steady state using a simple analytic relationship [*Ramaswamy et al.*, 2001].

3. Results and Discussion

3.1. Changes in Surface O_3 Metrics and Net Radiative Forcing

[11] Figure 1 shows the response of surface ozone (O_3^{srf}) to 20% decreases in anthropogenic precursor emissions. We show the daily maximum 8-hour O_3 averaged over the "O_3 season," which we define for each grid cell individually as the consecutive three-month period with highest average 8-hr. O_3 in the base simulation. The reduction in O_3^{srf} due to the decrease in CH_4 emissions is widespread globally, with the largest effects (>1 ppbv) in North Africa, the Middle East, and Europe (due to greater down-welling from the free troposphere and availability of NO_x) in agreement with previous results using different models or meteorology [*Fiore et al.*, 2002; *West and Fiore*, 2005; *West et al.*, 2006]. In contrast, the responses to changes in NO_x, NMVOCs and CO are concentrated near source regions,

with changes in background concentrations occurring mainly in the temperate Northern Hemisphere. In some locations (e.g., Northern Russia), NO_x reductions increase the modeled O_3^{srf} due to the decreased local destruction of O_3 by reaction with fresh NO emissions.

[12] Figure 2 shows the changes in CH_4 and O_3 concentrations resulting from reductions in emissions of each precursor. The 20% CH_4 reduction yields the greatest decrease in both the tropospheric O_3 burden (-6.7 Tg O_3; Figure 2b) and the global annual average O_3^{srf} (-0.61 ppbv; Figure 2c). Our estimated reduction of 0.12 Tg O_3 per Tg yr^{-1} change in CH_4 emissions is within the range of other values in the literature of 0.09 to 0.19 [*Prather et al.*, 2001; *Fiore et al.*, 2002; *Shindell et al.*, 2005; *West et al.*, 2006]. Changes in the O_3 burden also compare well for NO_x (0.29 Tg O_3 per Tg yr^{-1} (as NO_2), compared to 0.25 to 0.47 from other studies) [*Fiore et al.*, 2002; *Shindell et al.*, 2005; *Naik et al.*, 2005]. Finally, results for the CO (0.013 Tg O_3 per Tg yr^{-1}) and NMVOC reductions (0.12 Tg O_3 per Tg C yr^{-1}) agree well with those reported by *Fiore et al.* [2002] (0.014 and 0.14, respectively) (see auxiliary material).

[13] Figure 2c also includes changes in O_3^{srf} metrics relevant for air quality and human health, including the population-weighted daily maximum 8-hr. O_3, averaged over the whole year and over the O_3 season. The 20% NO_x reduction yields the greatest decrease in the population-weighted O_3^{srf} metrics, as the ΔO_3^{srf} is concentrated in populous source regions.

[14] In the United States, O_3^{srf} responds strongly to the 20% NO_x reduction, reflecting high regional NO_x emissions and NO_x-sensitive chemistry due to the high emissions of biogenic NMVOCs in the eastern US. In Europe, O_3^{srf} is particularly sensitive to changes in CH_4. The NMVOC emission reduction causes rather small changes in O_3^{srf}, in part because of large biogenic NMVOC emissions (the 20% anthropogenic reduction is only 1.0% of total NMVOC emissions). Note that the global model likely underestimates the effects of changes in NMVOC emissions on O_3 air quality in urban areas, particularly for the population-weighted metrics, due to its coarse resolution and lack of detail for highly reactive VOCs. For NO_x decreases, global models may either under- or overestimate O_3^{srf} changes in urban regions [*Liang and Jacobson*, 2000; *Karamchandani et al.*, 2002]. Local sensitivity to changes in emissions of NMVOCs and NO_x is best determined using a local or regional model which has been tested for local conditions.

[15] Figure 2c also shows a notable difference between the short-term and steady-state ΔO_3^{srf} metrics, due to long-term changes in CH_4. The 20% NO_x reduction causes long-term increases in the population-weighted O_3^{srf} metrics of about 0.2 ppbv, which counteract the short-term O_3^{srf} decreases (1.3 to 3.4 ppbv) by 6 to 14%. For the CO reduction, the short-term ΔO_3^{srf} (0.4 to 0.5 ppbv) is amplified by about 0.1 ppbv at steady state (a 16–21% increase). For NMVOCs, the additional long-term O_3^{srf} decrease is small (about 0.01 ppbv). These long-term changes in O_3^{srf} are globally widespread, following the spatial pattern of ΔO_3^{srf} from the CH_4 perturbation (Figure 1).

[16] Changes in net radiative forcing (RF_{net}) are shown in Figure 2d. The largest negative RF_{net} results from the CH_4 reduction, with 76% of the ΔRF_{net} from the decrease in CH_4 itself, and the remainder from the decrease in O_3. While reducing each precursor causes a negative forcing due to O_3, the NO_x reduction causes a larger positive CH_4 forcing, producing a positive global average ΔRF_{net}. This agrees with previous results showing that the ΔRF_{net} is positive for surface NO_x emission reductions in all world regions [*Fuglestvedt et al.*, 1999; *Berntsen et al.*, 2005; *Naik et al.*, 2005] (see auxiliary material for a comparison with other RF_{net} estimates).

[17] Changes in ozone precursor emissions also affect the concentrations of aerosols, through changes in oxidant chemistry. We find that the CH_4 reduction decreases the tropospheric burden of sulfate aerosol by 0.23%, in agreement with previous results [*West et al.*, 2006]. This decrease results from a competition between decreased heterogeneous sulfate production by hydrogen peroxide (-0.9 Tg yr^{-1}, or -1.1%) and increased gas-phase production by OH (+0.55 Tg yr^{-1}, or +2.5%). Decreases in other precursor emissions are likewise estimated to decrease the global sulfate burden: -0.16% when decreasing CO, -0.14% for NMVOCs, and -0.06% for NO_x. The global mean positive radiative forcing by the direct effect resulting from each of these decreases in sulfate are estimated to be less than $\sim$0.002 W m^{-2} (see auxiliary material), smaller than the forcings in Figure 2 but potentially important for local forcing [*Unger et al.*, 2006].

3.2. Change in RF_{net} Per Unit Change in O_3^{srf} Metrics

[18] We consider the ratio $\Delta RF_{net}/\Delta O_3^{srf}$ as an indicator of the effects on radiative forcing of actions to decrease metrics of O_3 air quality by a given amount. High positive values indicate that actions to reduce O_3^{srf} by one unit cause a large negative radiative forcing. In Figure 2e, the CH_4 emission reduction causes the largest decrease in RF_{net} per unit ΔO_3^{srf}, mainly due to the decreased forcing from CH_4 itself. Reductions in NO_x increase RF_{net}, causing a negative $\Delta RF_{net}/\Delta O_3^{srf}$. Abatement of CO emissions leads to a greater $\Delta RF_{net}/\Delta O_3^{srf}$ than for NMVOCs, as the ratio of the reduction in CH_4 to O_3 is greater for CO. We find that the order of $\Delta RF_{net}/\Delta O_3^{srf}$ (CH_4, CO, NMVOCs, NO_x) is the same for all ΔO_3^{srf} metrics considered, and we expect this order to be robust over model uncertainties as well. If ΔRF_{net} were evaluated after a short time period (<10 yr), however, the change in short-term O_3 forcing would dominate, and the order of the precursors would likely differ.

4. Conclusions

[19] We first consider the sensitivity of surface O_3 air quality and net radiative climate forcing to reductions in global O_3 precursor emissions. For O_3 air quality, the 20% reduction in anthropogenic NO_x emissions has the greatest effect on the population-weighted O_3^{srf} metrics analyzed, followed by CH_4, although the long lifetime of CH_4 delays realization of the O_3 decrease. The CH_4 reduction causes the greatest decrease in RF_{net}, mainly because of the direct reduction in CH_4 forcing, while NO_x emission reductions increase RF_{net}. Second, we present an indicator of the climate forcing resulting from actions to improve O_3 air quality ($\Delta RF_{net}/\Delta O_3^{srf}$). We find that of the means of decreasing O_3^{srf} metrics by one unit, abatement of CH_4 emissions best reduces radiative forcing.

[20] This research also demonstrates that the long-term changes in surface O_3 concentrations via changes in CH_4 are substantial for NO_x and CO reductions, and relevant for air quality management. While the 20% anthropogenic NO_x emission reduction decreases short-term O_3^{srf}, it also causes a long-term global increase of about 0.2 ppbv in population-weighted O_3^{srf} metrics, which counteracts the short-term decrease by 6 to 14%. For CO, the long-term O_3^{srf} reduction of 0.1 ppbv adds 16 to 21% to the short-term decrease, while the long-term O_3^{srf} reduction of the 20% NMVOC reduction is small (0.01 ppbv).

[21] We also find that reducing emissions of each precursor decreases the global sulfate aerosol burden (<0.3% for the 20% anthropogenic reductions). Future research should further address the effects of these emission changes on all relevant aerosol species (including secondary organics), and evaluate impacts on both surface air quality and global and regional aerosol radiative forcing. Further, while the methods presented here are convenient for considering short-term and steady-state changes, future research should conduct transient simulations with CH_4 modeled explicitly, to explore possible interactions between reductions in different species under future scenarios [*Dentener et al.*, 2005].

[22] In addition to the traditional focus of O_3 air quality management on local and regional scales, changes in precursor emissions also cause long-term changes in O_3^{srf} and radiative climate forcing on a global scale. Although the long-term effects of actions to manage O_3^{srf} in a single airshed are expected to be small, the cumulative effects of such actions globally are significant. Because of these long-term air quality and climate effects, therefore, it may be desirable to emphasize CH_4 abatement, and to increase the emphasis of O_3 air quality control on CO and NMVOC abatement. While the $\Delta RF_{net}/\Delta O_3^{srf}$ indicators presented here are useful in considering the climate effects of actions to improve air quality, we would ideally like to know the least-cost combination of emission reduction measures that would jointly achieve air quality and climate objectives [*West et al.*, 2004], and future work should combine our results with relevant control costs. In the case of CH_4, a large potential for low-cost and cost-saving emission controls has been identified, mainly in industrial sectors [*West and Fiore*, 2005].

[23] **Acknowledgments.** We thank H. Levy, P. Ginoux, and anonymous reviewers. This work was supported by the National Oceanic and Atmospheric Administration, and a National Aeronautics and Space Administration New Investigator Program grant to D.L.M.

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A. M. Fiore, L. W. Horowitz, and M. D. Schwarzkopf, NOAA Geophysical Fluid Dynamics Laboratory, 201 Forrestal Road, P.O. Box 308, Princeton, NJ 08542-0308, USA.

D. L. Mauzerall and V. Naik, Woodrow Wilson School of Public and International Affairs, Princeton University, Robertson Hall, Princeton, NJ 08544, USA.

J. J. West, Department of Environmental Sciences and Engineering, University of North Carolina, Chapel Hill, NC 27599, USA. (jwest@princeton.edu)