Linking ozone pollution and climate change: The case for controlling methane

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[1] Methane (CH₄) emission controls are found to be a powerful lever for reducing both global warming and air pollution via decreases in background tropospheric ozone (O₃). Reducing anthropogenic CH₄ emissions by 50% nearly halves the incidence of U.S. high-O₃ events and lowers global radiative forcing by 0.37 W m⁻² (0.30 W m⁻² from CH₄, 0.07 W m⁻² from O₃) in a 3-D model of tropospheric chemistry. A 2030 simulation based upon IPCC A1 emissions projections shows a longer and more intense U.S. O₃ pollution season despite domestic emission reductions, indicating that intercontinental transport and a rising O₃ background should be considered when setting air quality goals.


1. Introduction

[2] There is growing interest in linking air quality and climate change mitigation objectives in the design of emission control strategies. Tropospheric O₃ deserves particular attention as both the primary constituent of smog [National Research Council (NRC), 1991] and a significant greenhouse gas [Prather et al., 2001]. Ozone is produced in the troposphere by photochemical oxidation of volatile organic compounds (VOC) and carbon monoxide (CO) in the presence of nitrogen oxides (NOₓ). While O₃ production on global and regional scales is sensitive to NOₓ emissions from fossil fuel combustion [NRC, 1991; Wang and Jacob, 1998], reducing these emissions may increase greenhouse warming if the positive forcing from increased CH₄ concentrations offsets the negative forcing from decreased O₃ concentrations [Wild et al., 2001].

[3] Methane is a known major source of the tropospheric O₃ background, but is not generally considered a precursor to regional O₃ pollution episodes in surface air because of its long lifetime (8–9 years). Recent recognition that intercontinental transport may contribute to these pollution episodes [Jacob et al., 1999; Yienger et al., 2000; Wild and Akimoto, 2001; Fiore et al., 2002; Li et al., 2002] raises the profile of CH₄. The present-day U.S. O₃ standard is based upon a 0.08 ppmv (8-hour average), not to be exceeded more than three times per year. If this standard becomes more stringent, as it is in European countries (55–65 ppbv), the relative contribution of the background component to exceedances of the standard will increase. We show that reductions in CH₄ emissions deserve consideration as a means to meet air quality standards while simultaneously lessening radiative forcing.

2. Model Description

[4] We apply GEOS-CHEM (v4.16), a 3-D global model of tropospheric O₃-NOₓ-CO-VOC chemistry [Bey et al., 2001], to investigate the response of U.S. pollution episodes and global O₃ and CH₄ to (1) 50% reductions in various anthropogenic precursor emissions relative to a 1995 base year and (2) projected 2030 emissions from the IPCC A1 and B1 scenarios [Prather et al., 2001], which project relatively pessimistic and optimistic futures, respectively. All simulations were spun up for 6 months, long enough to remove the effects of initial O₃ concentrations on the results. Our simulations use assimilated observations of meteorological fields from NASA GEOS-1 with 20 vertical sigma layers and a 4° × 5° horizontal resolution; comparison with 2° × 2.5° resolution shows no significant bias [Fiore et al., 2002].

[5] The coarse resolution precludes the model from capturing the local O₃ maxima that determine compliance with the national O₃ standard. These maxima, however, typically occur under regionally stagnant conditions that are conducive to the formation of elevated O₃ levels spanning large spatial scales (~600,000 km²) [Logan, 1989] resolved by the model. During the summer of 1995, these stagnation episodes occurred more frequently than would be predicted by climatological averages [McNider et al., 1998]. We have previously shown that GEOS-CHEM captures these regional high-O₃ events, as well as the frequency distribution of O₃ at U.S. sites [Fiore et al., 2002]. We compare here simulated June–August daily afternoon (1–5 p.m. local time) mean O₃...
concentrations with surface measurements at EPA AIRS sites averaged onto the GEOS-CHEM grid in the eastern U.S. (east of 97.5°W where the network is most dense). The correlation coefficient (r) between observed and simulated daily afternoon concentrations is 0.68. The percentages of total grid-square afternoons (92 summer afternoons times 27 grid-squares) in excess of 80 ppbv, 70 ppbv, and 60 ppbv, are 5%, 15%, and 36%, respectively, in the observations; the corresponding percentages in the model are 1%, 10%, and 37%, respectively. In this study, we adopt a threshold of 70 ppbv as a metric for polluted conditions since the model provides good statistics for the occurrence of events above this threshold.

[6] The GEOS-CHEM emissions are described in detail by Bey et al. [2001] and Martin et al. [2002]. We classify biomass burning emissions as natural, following Fiore et al. [2002]. The anthropogenic contributions to total global emissions for NOx, CO, CH4, and NMVOC in the base case 1995 simulation (Table 1) are 65%, 50%, 60% and 12%, respectively. The CH4 concentration for 1995 is fixed at its uniform observed value of 1700 ppbv. This value implies a global emission of 570 Tg CH4 yr−1, computed from mass balance with the simulated global sink (oxidation by OH), plus a 4.9 ppbv yr−1 tropospheric accumulation. Following Prather et al. [2001], we view 60% of CH4 emissions as anthropogenic. We use the relationship between CH4 emissions and concentrations (assuming an OH feedback factor of 1.6) determined by Prather [1996] to translate a 50% decrease in anthropogenic CH4 emissions into a steady-state concentration of 1000 ppbv for the corresponding troposphere.

[7] Emissions for the A1 and B1 2030 scenarios were generated by scaling 1995 emissions by regional growth factors derived from emissions in the IMAGE socioeconomic model [IMAGE Team, 2001] for each anthropogenic source and biomass burning. The IMAGE model gives projections consistent with those of the IPCC reference models. Due to space restrictions, we focus here on the A1 scenario. Emissions increase globally (Table 1) from 1995 to 2030, but the distribution shifts. In the developed world (Europe, North America, Japan, etc.), anthropogenic NOx emissions decline by 10%, but they increase by 130% in the developing world (South America, Africa, Asia, etc.). U.S. anthropogenic emissions of O3 precursors decline by 20–40%. IMAGE projects a 43% increase in CH4 emissions from 1995 to 2030, which we apply to the CH4 concentration assuming constant OH. In the 2030 A1 simulation, CH4 actually increases by 31% (as implied by mass balance) because OH concentrations decrease. Thus our CH4 increase is conservative. The IPCC A1 scenario does not include aircraft NOx emissions; we use projections from the Environmental Defense Fund [Henderson et al., 1999] to determine a growth factor of 2.26.

### Table 1. Annual Emissions and Growth Factors in GEOS-CHEM

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<tr>
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<tbody>
<tr>
<td>NOx</td>
<td>27</td>
<td>15</td>
<td>1.8</td>
<td>1.0</td>
</tr>
<tr>
<td>CO</td>
<td>580</td>
<td>630</td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>CH4</td>
<td>340</td>
<td>230</td>
<td>1.3</td>
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### Table 2. Impacts of Perturbations to Anthropogenic Emissions

<table>
<thead>
<tr>
<th>Perturbation</th>
<th>Base Case 1995</th>
<th>50% CH4</th>
<th>50% NOx</th>
<th>50% VOC</th>
<th>50% NOx &amp; VOC</th>
<th>50% CO</th>
<th>50% All</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tropospheric O3 burden (Tg)</td>
<td>321</td>
<td>294</td>
<td>300</td>
<td>317</td>
<td>297</td>
<td>317</td>
<td>269</td>
</tr>
<tr>
<td>Global CH4 conc. (ppbv)</td>
<td>1700</td>
<td>1000</td>
<td>1867</td>
<td>1685</td>
<td>1846</td>
<td>1643</td>
<td>1040</td>
</tr>
<tr>
<td>OPE</td>
<td>33</td>
<td>30</td>
<td>43</td>
<td>32</td>
<td>42</td>
<td>32</td>
<td>38</td>
</tr>
<tr>
<td>CH4 lifetime</td>
<td>8.5</td>
<td>7.2</td>
<td>9.4</td>
<td>8.5</td>
<td>9.3</td>
<td>8.3</td>
<td>7.5</td>
</tr>
</tbody>
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### Footnotes

8 Emissions from biomass burning are considered natural [Fiore et al., 2002].
9 Concentrations are specified for the base case, A1, and 50% CH4 simulations. For the other simulations, concentrations are derived from the model computed OH concentrations by assuming the same CH4 source as in the 1995 base case simulation.
10 Ozone Production Efficiency (OPE) is defined as the ratio of O3 (actually odd oxygen) production to total NOx emissions, following the procedure used by Wang and Jacob [1996].
11 Total atmospheric burden of CH4 (4760 Tg in GEOS-CHEM) divided by tropospheric loss of CH4 by reaction with OH.
12 Background is defined as O3 produced outside of the North American boundary layer [Fiore et al., 2002] (see text).
burden (Table 2) than 50% reductions in anthropogenic NOx emissions. This might appear to be inconsistent with the O3 source inferred from scaling global NOx emissions by the OPEs in Table 2. But, anthropogenic NOx emissions have low OPEs due to chemical nonlinearity and titration by NOx [Liang et al., 1998; Kasishtia et al., 1998]. Thus, anthropogenic NOx emissions are less effective than natural (in particular from lightning) in contributing to the global O3 budget, whereas the homogeneity of CH4 permits anthropogenic and natural CH4 emissions to be equally effective.

[10] Climatic implications of an atmospheric perturbation can be assessed using the standard concept of radiative forcing [Ramaswamy et al., 2001] which describes the instantaneous global change in the radiative balance of the Earth system resulting from the perturbation. We calculate the change in radiative forcing from CH4 directly from the global change in concentration [Ramaswamy et al., 2001], while for O3 we use a relationship of 0.034 W m$^{-2}$ per Dobson Unit change in the mean tropospheric column [Mickley et al., 1999; Ramaswamy et al., 2001]. Results are shown in the top panel of Figure 1. A 50% reduction in anthropogenic CH4 emissions yields the largest decrease in radiative forcing ($-0.37$ W m$^{-2}$, $-0.30$ W m$^{-2}$ from CH4 and $-0.07$ W m$^{-2}$ from O3). The radiative effect of 50% reductions in anthropogenic NOx emissions is neutral, as the positive forcing due to decreased O3 is balanced by the negative forcing from increased CH4 [Fuglestvedt et al., 1999]. Both 2030 simulations show positive forcing from 1995 to 2030 (A1: 0.44 W m$^{-2}$, B1: 0.19 W m$^{-2}$).

### Figure 1. Response of radiative forcing and U.S. air quality to (1) 50% global decreases in anthropogenic emissions relative to 1995, (2) IPCC A1 and B1 emissions projections for 2030. (upper panel) Global radiative forcing relative to 1995 as calculated from changes in CH4 and O3 concentrations. (lower panel) Number of model grid-square days in the U.S. in summer with mean afternoon (1–5 p.m.) surface O3 concentrations in excess of 70 ppbv.

4. Impacts on U.S. O3 Pollution

[11] We next examine how summertime (June–August) surface air quality over the U.S. responds to the perturbed emissions, focusing on afternoons (1–5 p.m. local time) when O3 concentrations typically peak. We define background O3 as that produced outside of the North American boundary layer (surface to 700 hPa). It is diagnosed by applying production and loss rates archived from the full chemical simulation to drive an off-line simulation where O3 is divided into individual tagged tracers which are produced in different regions of the atmosphere [Fiore et al., 2002]. Table 2 shows mean and background O3 concentrations simulated on summer afternoons in surface air over the U.S. Summer afternoon O3 concentrations respond most strongly to the 50% reductions in anthropogenic NOx emissions, but reductions in CH4 emissions also have a large effect. The decreases in NOx and CH4 emissions, however, are equally effective at lowering background O3 concentrations (2 ppbv in the mean). Decreases in CO or VOC emissions have little impact. For the A1 2030 simulation, mean afternoon surface O3 increases by 4 ppbv while background concentrations rise by 6 ppbv. Even in the more optimistic B1 scenario, higher global CH4 emissions contribute to elevating U.S. background O3 levels by 2 ppbv, partially offsetting air quality gains achieved via domestic emissions controls.

[12] We use a threshold of 70 ppbv as a metric to gauge changes in the frequency of O3 pollution events in our simulations. Figure 1 (bottom panel) shows the number of U.S. grid-square days in June–August 1995 where simulated afternoon average (1–5 p.m. local time) O3 levels exceed 70 ppbv. Reducing anthropogenic NOx emissions by 50% nearly eliminates the occurrence of grid-square days in excess of 70 ppbv (Figure 1). Because of subgrid variability in O3, this result does not mean that local exceedances of 70 ppbv would be as drastically reduced, but it does point to significant improvement in air quality. When anthropogenic CH4 emissions are reduced by 50%, the incidence of O3 concentrations in excess of 70 ppbv (Figure 1) declines by 45% (for an 80 ppbv threshold that statistic is 54%).

[13] The U.S. has aggressive emission controls to abate future O3 pollution. Although fossil fuel emissions in the U.S. for NOx, CO, and VOC decline in the IPCC A1 2030 scenario relative to 1995 by 27%, 45%, and 30%, respectively, we find that the number of grid-square summer days over the U.S. with O3 > 70 ppbv increases relative to 1995. Efforts to improve U.S. air quality are thus thwarted by the rise in global background O3 levels due to increased emissions outside U.S. borders. These results are consistent with the modeling study of Collins et al. [2000] who found...
that European efforts to improve air quality via domestic emissions reductions may be offset by a rise in emissions from developing nations by 2015. These simulations underscore the need to consider regional air quality in a global context.

[14] Another adverse impact of rising global emissions on U.S. air quality is a longer U.S. O₃ pollution season, as diagnosed by exceedances of a 70 ppbv threshold. Intercontinental transport makes a larger contribution to U.S. O₃ pollution in spring and fall than in summer because of the longer O₃ lifetime [Jacob et al., 1999]. Figure 2 shows the larger numbers of exceedances of the 70 ppbv threshold in March through November in the A1 2030 simulation; results are similar for an 80 ppbv O₃ threshold.

5. Conclusions

[15] Our global 3-D model analysis shows that reducing CH₄ emissions enables a simultaneous pursuit of O₃ air quality and climate change mitigation objectives. Whereas reductions in NOₓ emissions achieve localized decreases in surface O₃ concentrations, reductions in CH₄ emissions lower the global O₃ background and improve surface air quality everywhere. Simulation of a 2030 (IPCC A1) scenario where anthropogenic U.S. emissions of O₃ precursors decrease but global emissions (including CH₄) increase indicates a greater incidence of O₃ pollution episodes and a longer U.S. O₃ season, stressing the need for a global perspective in the design of future regional pollution control strategies.

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