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

The Harvard community has made this article openly available. Please share how this access benefits you. Your story matters

Citation

Published Version
doi:10.1029/2002GL015601

Citable link
http://nrs.harvard.edu/urn-3:HUL.InstRepos:14121777

Terms of Use
This article was downloaded from Harvard University’s DASH repository, and is made available under the terms and conditions applicable to Other Posted Material, as set forth at http://nrs.harvard.edu/urn-3:HUL.InstRepos:dash.current.terms-of-use#LAA
Linking ozone pollution and climate change: 
The case for controlling methane

Arlene M. Fiore, Daniel J. Jacob, and Brendan D. Field
Department of Earth and Planetary Sciences and Division of Engineering and Applied Sciences, Harvard University, Cambridge, MA, USA

David G. Streets and Sunetna D. Fernandes
Argonne National Laboratory, Argonne, IL, USA

Carey Jang
USEPA/OAQPS MC: D243-01, RTP, NC, USA

Received 4 June 2002; revised 8 August 2002; accepted 13 August 2002; published 8 October 2002.

[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. INDEX TERMS: 0325 Atmospheric Composition and Structure: Evolution of the atmosphere; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry. Citation: Fiore, A. M., D. J. Jacob, B. D. Field, D. G. Streets, S. D. Fernandes, and C. Jang, Linking ozone pollution and climate change: The case for controlling methane. Geophys. Res. Lett., 29(19), 1919, doi:10.1029/2002GL015601, 2002.

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; Ytenger 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 ppbv (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...
Table 1. Annual Emissions and Growth Factors in GEOS-CHEM

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>N</td>
<td>A</td>
<td>N</td>
</tr>
<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>VOC</td>
<td>58</td>
<td>450</td>
<td>1.6</td>
<td>1.0</td>
</tr>
<tr>
<td>CH4</td>
<td>340</td>
<td>230</td>
<td>1.3</td>
<td>1.3</td>
</tr>
</tbody>
</table>

*a A = anthropogenic; N = natural (includes biomass burning). Emissions are in units of Tg N yr⁻¹ for NOx, Tg CO for CO, Tg C for VOC and Tg CH4 for CH4.


*c Nonmethane volatile organic compounds; natural emissions include biogenic isoprene (404 Tg C), propene (12 Tg C), acetone (15 Tg C), biomass burning.

*d The contribution from U.S. emissions is not resolved because a uniform mixing ratio is applied (see text).

*e Calculated as described in the text.

Table 2. Impacts of Perturbations to Anthropogenic Emissions

<table>
<thead>
<tr>
<th>Selected Diagnostics</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>
<th>A1 2030</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>
<td>394</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>
<td>2431</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>
<td>27</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>
<td>9.3</td>
</tr>
</tbody>
</table>

Mean U.S. summer afternoon surface O3 (ppbv)

| Mean | 51 | 48 | 42 | 50 | 41 | 50 | 38 | 55 |
| Background | 23 | 21 | 21 | 23 | 21 | 23 | 18 | 29 |

*a Emissions from biomass burning are considered natural [Fiore et al., 2002].

*b 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.

*c 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 [1998].

*d Total atmospheric burden of CH4 (4760 Tg in GEOS-CHEM) divided by tropospheric loss of CH4 by reaction with OH.

*e Background is defined as O3 produced outside of the North American boundary layer [Fiore et al., 2002] (see text).
4. Impacts on U.S. O$_3$ Pollution

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 O$_3$ concentrations typically peak. We define background O$_3$ 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 O$_3$ 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 O$_3$ concentrations simulated on summer afternoons in surface air over the U.S. Summer afternoon O$_3$ concentrations respond most strongly to the 50% reductions in anthropogenic NO$_x$ emissions, but reductions in CH$_4$ emissions also have a large effect. The decreases in NO$_x$ and CH$_4$ emissions, however, are equally effective at lowering background O$_3$ concentrations (2 ppbv in the mean). Decreases in CO or VOC emissions have little impact. For the A1 2030 simulation, mean afternoon surface O$_3$ increases by 4 ppbv while background concentrations rise by 6 ppbv. Even in the more optimistic B1 scenario, higher global CH$_4$ emissions contribute to elevating U.S. background O$_3$ levels by 2 ppbv, partially offsetting air quality gains achieved via domestic emissions controls.

We use a threshold of 70 ppbv as a metric to gauge changes in the frequency of O$_3$ 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) O$_3$ levels exceed 70 ppbv. Reducing anthropogenic NO$_x$ emissions by 50% nearly eliminates the occurrence of grid-square days in excess of 70 ppbv (Figure 1). Because of subgrid variability in O$_3$, 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 CH$_4$ emissions are reduced by 50%, the incidence of O$_3$ concentrations in excess of 70 ppbv (Figure 1) declines by 45% (for an 80 ppbv threshold that statistic is 54%).

The U.S. has aggressive emission controls to abate future O$_3$ pollution. Although fossil fuel emissions in the U.S. for NO$_x$, 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 O$_3$ > 70 ppbv increases relative to 1995. Efforts to improve U.S. air quality are thus thwarted by the rise in global background O$_3$ 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.

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.

Figure 2. Number of U.S. model grid-square days per month with afternoon (1–5 p.m.) O₃ concentrations in surface air above a 70 ppbv threshold for the 1995 base case (white bars) and 2030 A1 (black bars) simulations. The A1 simulation reveals a longer O₃ pollution season. Results are similar for an 80 ppbv O₃ threshold.

5. Conclusions

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.

Acknowledgments. The authors would like to thank Loretta Mickley and Mat Evans for helpful conversations, and to acknowledge the support of the U.S. EPA Office of Air Quality and Planning Standards, the National Science Foundation, and the Atmospheric Chemistry Modeling and Analysis Program of NASA.