Causes of Changing Air Quality and Climate: Current Understanding & Relevant Issues

Michael J. Prather
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What species drive climate change and air quality on a global scale?

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What is the coupling between urban AQ and climate change?

Johnson et al / Hogrefe et al(?) - climate change alters trop O3 & AQ exceedances
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IPCC/TAR Radiative Forcing 1750 → 2000
IPCC/TAR SRES A2 → 2100
IPCC/TAR SRES A2 → 2100
Processes Controlling Tropospheric $O_3$

Strat.-Trop. Exchange

Emissions

Deposition

(from O. Wild)
How do *criteria pollutants* interact with *greenhouse gases*?

CO, VOC, NO$_x$ (=NO+NO$_2$), & CH$_4$ control

Tropospheric Chemistry (OH, HO$_2$)

which is the sink for CH$_4$ & HFCs; the source for O$_3$
$CH_4$ feedback on its lifetime

$Isaksen \ & \ Hov, \ 1987$

$Prather, \ 1994$

$IPCC \ SAR \ (multi-model \ assessment), \ 1995$
CH4 self-feedback, 2001

Changes in atmospheric composition and chemistry over the past century have affected, and those projected into the future will affect, the lifetimes of many greenhouse gases and thus alter the climate forcing of anthropogenic emissions:

The atmospheric lifetime relates emissions of a component to its atmospheric burden. In some cases, for instance for methane, a change in emissions perturbs the chemistry and thus the corresponding lifetime. The CH4 feedback effect amplifies the climate forcing of an addition of CH4 to the current atmosphere by lengthening the perturbation lifetime relative to the global atmospheric lifetime of CH4 by a factor of 1.4. This earlier finding is corroborated here by new model studies that also predict only small changes in this CH4 feedback for the different scenarios projected to year 2100. Another feedback has been identified for the addition of N2O to the atmosphere; it is associated with stratospheric O3 chemistry and shortens the perturbation lifetime relative to the global atmospheric lifetime of N2O by about 5%.
CO becomes an indirect greenhouse gas

- Tropical bio-burn
- NH industrial

3D CTM: add CO

CH₄ increase

O₃ increase

CO long-term

O₃ short-term
$H_2$ is likewise an indirect greenhouse gas

![Diagram showing the relationship between $H_2$, OH, CH$_4$, and O$_3$.]

- Long-term CH$_4$ increase
- Short-term O$_3$ increase

3D CTM: add $H_2$
- Tropical bio-burn
- NH industrial
Time scales in atmospheric chemistry: Theory, GWPs for CH$_4$ and CO, and runaway growth

Michael J. Prather
Earth System Science, University of California at Irvine

Abstract. Atmospheric CH$_4$ perturbations, caused directly by CH$_4$ emissions or indirectly by those of CO are enhanced by chemical feedbacks. They can be diagnosed in terms of the natural modes of atmospheric chemistry that are general solutions of the continuity equations. Each mode is a pattern in the global distribution of all chemical species, and each has a single time-constant that accurately describes its exponential decay about a given atmospheric state. This mathematical theory extends earlier work and is general for 2-D and 3-D chemistry-transport models. A formal proof relates the steady-state distribution and its lifetime to the integral of the true time-dependent response (properly included in the recent IPCC assessment). Changes in CO are also known to perturb CH$_4$; however, the impact of CO emissions on climate has not been formally assessed in part because the short lifetime of CO (months) relative to that of CH$_4$ (decades) was believed to limit the integrated impact. Using the IPCC model studies, this theory predicts that adding 5 CO molecules to today's atmosphere is equivalent to adding 1 CH$_4$ molecule with the same decadal duration as direct CH$_4$ addition. Extrapolating these results, CH$_4$ sources would have to triple before runaway growth, when CH$_4$ emissions exceed the oxidizing capacity of the troposphere.
Executive Summary

Two important new findings since the IPCC WGI Second Assessment Report (IPCC, 1996) (hereafter SAR) demonstrate the importance of atmospheric chemistry in controlling greenhouse gases:

Currently, tropospheric ozone ($O_3$) is the third most important greenhouse gas after carbon dioxide ($CO_2$) and methane ($CH_4$). It is a product of photochemistry, and its future abundance is controlled primarily by emissions of $CH_4$, carbon monoxide ($CO$), nitrogen oxides ($NO_x$), and volatile organic compounds (VOC). There is now greater confidence in the model assessment of the increase in tropospheric $O_3$ since the pre-industrial period, which amounts to 30% when globally averaged, as well as the response to future emissions. For scenarios in which the $CH_4$ abundance doubles and anthropogenic CO and $NO_x$ emissions triple, the tropospheric $O_3$ abundance is predicted to increase by an additional 50% above today’s abundance.

$CO$ is identified as an important indirect greenhouse gas. An addition of $CO$ to the atmosphere perturbs the OH-$CH_4$-$O_3$ chemistry. Model calculations indicate that the emission of 100 Mt of $CO$ stimulates an atmospheric chemistry perturbation that is equivalent to direct emission of about 5 Mt of $CH_4$. 
NOx identified as an indirect greenhouse gas

NOx proposed as indirect greenhouse gas
(Shine, Derwent, et al, 1st IPCC Assessment Report)

Rejected as too far ahead of its time
(1992 IPCC Interim Report)

Post SAR, explicit in IPCC Aviation Assessment, NOx is indirect greenhouse gas.
NOx

O3

OH

CH4

O3

regional

(Wild, Prather, Akimoto, 2001)
Integrated Radiative Forcing (CH$_4$ & trop O$_3$) from 0.5 Tg-N as global fossil fuel

- Short-term O$_3$ increase
- Regional heating
- Long-term CH$_4$ decrease
- Global cooling

Year of Run

mW m$^{-2}$ yr

Emissions Period

0 10 20 30 40 50
**NOx** is an *indirect greenhouse gas* (Wild, Prather, Akimoto, 2001)

0.5 Tg-N of NOx: tropical, high-altitude emissions have greatest impact

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**Diagram:**
- **Increased trop-O₃ warming →**
- **Decreased CH₄ cooling →**

- **RF from long-lived mode (mW m⁻² yr):**
  - Europe
  - N. Am.
  - Industrial
  - E. Asia
  - Biomass
  - Surface NO
  - Tropics
- **RF from short-lived modes (mW m⁻² yr):**
  - Industrial NO + CO (+10 Tg-CO)
  - Aircraft NO
  - Lightning NO
Processes Controlling Tropospheric \( O_3 \)

- NO, NO\(_2\), OH, HO\(_2\), RO\(_2\)
- CO, O\(_3\)
- O\(_3\)
- H\(_2\)O

Deposition

Strat.-Trop. Exchange

Emissions

NMHCs, CH\(_4\), CO

OH

HO\(_2\), RO\(_2\)

O\(_3\)

CO, O\(_3\)

O\(_3\)

OH

HO\(_2\)

(from O. Wild)
Processes Controlling Tropospheric $O_3$

(from O. Wild)
Carmichael, G. et al., ....

Early work on aerosol-photochemistry coupling, but on regional scale, no integration of global climate impacts.
We evaluate the sensitivity of tropospheric OH, O₃, and O₃ precursors to photochemical effects of aerosols not usually included in global models: (1) aerosol scattering and absorption of ultraviolet radiation (*via Fast-J*), and (2) reactive uptake of HO₂, NO₂, and NO₃. ... Aerosols decrease the O₃→O(¹D) photolysis frequency by 5-20% at the surface throughout the Northern Hemisphere (largely due to mineral dust) and by a factor of 2 in biomass burning regions (largely due to black carbon). Aerosol uptake of HO₂ ... **Annual mean OH concentrations decrease by 9% globally and by 5-35% in the boundary layer over the Northern Hemisphere.** Simulated CO increases by 5-15 ppbv in the remote Northern Hemisphere, improving agreement with observations. Simulated boundary-layer O₃ decreases by 15-45 ppbv over India during the biomass burning season in March, and by 5-9 ppbv over northern Europe in August, again improving comparison with observations. We find that particulate matter controls would increase surface O₃ over Europe and other industrial regions.

*Figure 3:* Sensitivity to aerosols of the O₃→O(¹D) photolysis frequency (*J*(O(¹D))) in surface air. Values are monthly-mean model results for March and August 1997.

Aerosols affect the global budgets of O$_3$, OH, and CH$_4$ in part through their alteration of photolysis rates and in part through their direct chemical interactions with gases (i.e., “heterogeneous chemistry”). ... *Globally averaged, the impact of aerosols on photolysis alone is to increase tropospheric O$_3$ by 0.63 Dobson units and increase tropospheric CH$_4$ by 130 ppb (via tropospheric OH decreases of 8%).* These greenhouse gas increases lead to an aerosol indirect effect (counting both natural and anthropogenic aerosols) of +0.08 W/m$^2$. ... The predominant impact is due to the aerosols over land; aerosols over the ocean contribute less than a third to globally integrated changes.
Bian et al, 2003 –

Aerosol-O$_3$ photolytic coupling gives both increases and decreases in O$_3$

Figure 3. Perturbations to O$_3$ (ppb) by global aerosol at six atmospheric layers in January and July.

A unified tropospheric chemistry-aerosol model has been developed within the Goddard Institute for Space Studies general circulation model (GCM). The model includes a detailed simulation of tropospheric ozone-NOx-hydrocarbon chemistry as well as aerosols and aerosol precursors. Predicted aerosol species include sulfate, nitrate, ammonium, black carbon, primary organic carbon, and secondary organic carbon. The partitioning of ammonia and nitrate between gas and aerosol phases is determined by on-line thermodynamic equilibrium, and the formation of secondary organic aerosols is based on equilibrium partitioning and experimentally determined parameters. Two-way coupling between aerosols and chemistry provides consistent chemical fields for aerosol dynamics and aerosol mass for heterogeneous processes and calculations of gas-phase photolysis rates (*via Fast-J*). ... As a result of the coupling between chemistry and aerosols, global burdens of both gas-phase and aerosol species are predicted to respond non-linearly to changing emissions of NOx, NH3, and sulfur.
**Effective Reactive Pollutant contribution to the CH$_4$ budget today (based on TAR 2001, +...)**

<table>
<thead>
<tr>
<th>Pollutant</th>
<th>Emission (Tg/ch4)/y</th>
<th>Impact on CH$_4$ (Tg/ch4)/y</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH$_4$</td>
<td>320</td>
<td>= direct emission</td>
</tr>
<tr>
<td>NOx</td>
<td>40</td>
<td>⇔ - 80</td>
</tr>
<tr>
<td>CO</td>
<td>1000</td>
<td>⇔ + 50</td>
</tr>
<tr>
<td>VOC</td>
<td>250</td>
<td>⇔ + 30</td>
</tr>
<tr>
<td>aerosols</td>
<td></td>
<td>⇔ +?40</td>
</tr>
</tbody>
</table>
RF from Tropospheric O₃ (TAR)

Mean Tropospheric O₃ Change: 1750 ⇒ 2000

+9 DU
likely range +6 to +13 DU

14 ppb
(7 ppb SH & 21 ppb NH)
(??? surface)

0.36 W m⁻²
(??? +0.8 W m⁻²)
Trends in free tropospheric O₃

\[ \text{observed} \ 25 \text{ to } 34 \text{ DU} \]

\[ \text{modeled} \ \triangle \text{CH}_4 \ +4.6 \text{ DU} \]

\[ \text{cause} \ \triangle \text{NO}_X \ +4.1 \]

\[ \triangle \text{CO} \ +1.2 \]

\[ \triangle \text{VOC} \ +0.5 \]
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Role of climate feedback on methane and ozone studied with a coupled Ocean-Atmosphere-Chemistry model.

C. E. Johnson, D. S. Stevenson¹, W. J. Collins², and R. G. Derwent²
Met Office, Hadley Centre for Climate Prediction and Research, UK

with global warming:
► H₂O increases
► O₃ loss increases
► OH increases
► CH₄ decreases
Linking ozone pollution and climate change:
The case for controlling methane

Arlene M. Fiore, Daniel J. Jacob, a
Department of Earth and Planetary Sciences and
Harvard University, Cambridge, MA, USA

David G. Streets and Suneeta D. Fe
Argonne National Laboratory, Argonne, IL, USA

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

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
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 Suneeta D. Fernandes
Argonne National Laboratory, Argonne, IL, USA

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

[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
NOx (and $O_3$ production) stays close to sources or does it?


Seasonal mean tropospheric NO$_2$ columns for September 1996 – August 1997.
Chemical Production and Export of $O_3$ (= climate forcing)

Free Troposphere

Polluted PBL

Distant PBL

$O_3$, CO, NO$_y$

$P(O_3)$ Fast
$L(O_3)$ Fast
$\tau(O_3)$ Short

$P(O_3)$ Slow
$L(O_3)$ Slow
$\tau(O_3)$ Long

$P(O_3)$ Slow
$L(O_3)$ Moderate
$\tau(O_3)$ Short
Look at regional and global O$_3$ from a single day’s emissions over Shanghai in March 2001.

**March 12:** Sunny, high pressure = **Cook-then-mix**

**March 16:** Heavily overcast = **Mix-then-cook**

Regional production different, global production similar, but evolution quite different.
**Additional Ozone Burden**

- Added $O_3$ burden from a single day's emissions:
  - over **Region** (3-day integral: air quality)
  - over **Globe** (1-month integral: climate)
- Global impact smaller when regional ozone impact large
  - Tokyo has greater climate impact than Shanghai
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What are we concerned about?
IPCC TAR 2001

**tropospheric O₃**

what about surface O₃?

**lifetime of CH₄**
IPCC (2001). “Changes projected in the SRES A2 and A1FI scenarios would degrade air quality over much of the globe by increasing background levels of O₃. In northern mid-latitudes during summer, the zonal average increases near the surface are about 30 ppb or more, raising background levels to nearly 80 ppb, threatening attainment of air quality standards over most metropolitan and even rural regions, and compromising crop and forest productivity. This problem reaches across continental boundaries since emissions of NOx influence photochemistry on a hemispheric scale.”
Tar/Sres:

Anthropogenic Impact from Reactive Pollutants 1750 – 2000 – 2100 (A2x)

\[ \text{Trop-OH} \]

<table>
<thead>
<tr>
<th></th>
<th>1750</th>
<th>2000</th>
<th>2100 (A2x)</th>
</tr>
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<tbody>
<tr>
<td>( \text{CH}_4 )</td>
<td>-29</td>
<td>-29</td>
<td>%</td>
</tr>
<tr>
<td>( \text{NO}_{x}^* )</td>
<td>+14</td>
<td>+33</td>
<td>%</td>
</tr>
<tr>
<td>( \text{CO} )</td>
<td>-11</td>
<td>-15</td>
<td>%</td>
</tr>
<tr>
<td>( \text{VOC} )</td>
<td>-5</td>
<td>-6</td>
<td>%</td>
</tr>
<tr>
<td>aerosols</td>
<td>??</td>
<td></td>
<td></td>
</tr>
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</table>

\[ \text{Trop-O}_3 \]

<table>
<thead>
<tr>
<th></th>
<th>1750</th>
<th>2000</th>
<th>2100 (A2x)</th>
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</thead>
<tbody>
<tr>
<td>( \text{CH}_4 )</td>
<td>+4.6</td>
<td>+4.5</td>
<td></td>
</tr>
<tr>
<td>( \text{NO}_{x}^* )</td>
<td>+4.1</td>
<td>+9.7</td>
<td></td>
</tr>
<tr>
<td>( \text{CO} )</td>
<td>+1.2</td>
<td>+1.6</td>
<td></td>
</tr>
<tr>
<td>( \text{VOC} )</td>
<td>+0.5</td>
<td>+0.7</td>
<td></td>
</tr>
<tr>
<td>total</td>
<td>+10.4</td>
<td>+16.5</td>
<td>DU</td>
</tr>
</tbody>
</table>
non-SRES futures

Trends of measured climate forcing agents

James E. Hansen*† and Makiko Sato*†

The growth rate of climate forcing by measured greenhouse gases peaked near 1980 at almost 5 W/m² per century. This growth rate has since declined to ~3 W/m² per century, largely because of cooperative international actions. We argue that trends can be reduced to the level needed for the moderate “alternative” climate scenario (~2 W/m² per century for the next 50 years) by means of concerted actions that have other benefits, but the forcing reductions are not automatic “co-benefits” of actions that slow CO₂ emissions. Current trends of climate forcings by aerosols remain very uncertain. Nevertheless, practical constraints on changes in emission levels suggest that global warming at a rate +0.15 ± 0.05°C per decade will occur over the next several decades.

Fig. 7. Climate forcing scenario for 2000–2050 that yields a forcing of 0.85 W/m² (colored bars), including small forcings from stratospheric ozone recovery and trace gases (Table 1).
non-SRES futures

Climate forcings in Goddard Institute for Space Studies SI2000 simulations

J. Hansen,¹ M. Sato,¹,² L. Nazarenko,¹,² R. Ruedy,¹,³ A. Lacis,¹ D. Koch,¹,⁴ I. Tegen,⁵ T. Hall,¹,⁶ D. Shindell,¹ B. Santer,⁷ P. Stone,⁸ T. Novakov,⁹ L. Thomason,¹⁰ R. Wang,¹¹ Y. Wang,¹² D. Jacob,¹³ S. Hollandsworth,¹⁴ L. Bishop,¹⁵ J. Logan,¹³ A. Thompson,¹⁴ R. Stolarski,¹⁴ J. Lean,¹⁶ R. Willson,² S. Levitus,¹⁷ J. Antonov,¹⁷ N. Rayner,¹⁸ D. Parker,¹⁸ and J. Christy¹⁹
non-SRES futures

The impact of air pollutant and methane emission controls on tropospheric ozone and radiative forcing: CTM calculations for the period 1990–2030

F. Dentener¹, D. Stevenson², J. Cofala³, R. Mechler³, M. Amann³, P. Bergamaschi¹, F. Raes¹, and R. Derwent¹
The “Current Legislation” (CLE) scenario reflects the current perspectives of individual countries on future economic development and takes the anticipated effects of presently decided emission control legislation in the individual countries into account.

The “Maximum technically Feasible Reduction” (MFR) scenario considers emission reductions offered by full implementation of the presently available emission control technologies, while maintaining the projected levels of anthropogenic activities.

Whereas the resulting projections of methane emissions lie within the range suggested by other greenhouse gas projections, the recent pollution control legislation of many Asian countries, requiring introduction of catalytic converters for vehicles, leads to significantly lower growth in emissions of the air pollutants NOx, NMVOC and CO than in SRES.

These new scenarios form the core the new IPCC AR4 atmospheric chemistry studies (Stevenson and Dentener, ACCENTworkshop, Jan 2005, Oslo).
**NO$_x$ emissions**

![Graph showing NO$_x$ emissions]

*Fig. 1.* Projected development of IIASA anthropogenic NO$_x$ emissions by SRES world region (Tg NO$_2$yr$^{-1}$).
CO emissions

Fig. 2. Projected development of IIASA anthropogenic CO emissions by SRES world region (Tg CO yr$^{-1}$).
CH$_4$ emissions

**Fig. 3.** Projected development of IIASA anthropogenic CH$_4$ emissions by SRES region (Tg CH$_4$ yr$^{-1}$).
The impact of air pollutant and methane emission controls on tropospheric ozone and radiative forcing: CTM calculations for the period 1990–2030

F. Dentener\textsuperscript{1}, D. Stevenson\textsuperscript{2}, J. Cofala\textsuperscript{3}, R. Mechler\textsuperscript{3}, M. Amann\textsuperscript{3}, P. Bergamaschi\textsuperscript{1}, F. Raes\textsuperscript{1}, and R. Derwent\textsuperscript{4}

Table 3. Radiative Forcings calculated by TM3 and STOCHEM comparing the 2020s with the 1990s. Other: emissions of NO\textsubscript{x}-CO-NMVOC.

<table>
<thead>
<tr>
<th></th>
<th>(O_3)</th>
<th>(CH_4)</th>
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<tbody>
<tr>
<td>CLE TM3</td>
<td>0.075</td>
<td>0.167</td>
</tr>
<tr>
<td>CLE STOCHEM</td>
<td>0.041</td>
<td>0.125</td>
</tr>
<tr>
<td>MFR TM3</td>
<td>-0.073</td>
<td>0.004</td>
</tr>
<tr>
<td>MFR STOCHEM</td>
<td>-0.072</td>
<td>0.003</td>
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<tr>
<td>MFR-CH4</td>
<td>0.029</td>
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<tr>
<td>MFR-pol</td>
<td>-0.030</td>
<td>0.221</td>
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<tr>
<td>IPCC-TAR B1-A1FI 2000–2030</td>
<td>0.08–0.31</td>
<td>0.06–0.16</td>
</tr>
</tbody>
</table>
Air Pollution as Climate Forcing: A Second Workshop
April 4-6, 2005 at East-West Center, Honolulu

Mrs. Nimmers uses localism to combat globalism.