

## Fresh air in the 21st century?

Michael Prather,<sup>1</sup> Michael Gauss,<sup>2</sup> Terje Berntsen,<sup>2</sup> Ivar Isaksen,<sup>2</sup> Jostein Sundet,<sup>2</sup> Isabelle Bey,<sup>3</sup> Guy Brasseur,<sup>4</sup> Frank Dentener,<sup>5</sup> Richard Derwent,<sup>6</sup> David Stevenson,<sup>6</sup> Lee Grenfell,<sup>7</sup> Didier Hauglustaine,<sup>8</sup> Larry Horowitz,<sup>9</sup> Daniel Jacob,<sup>10</sup> Loretta Mickley,<sup>10</sup> Mark Lawrence,<sup>11</sup> Rolf von Kuhlmann,<sup>11</sup> Jean-Francois Muller,<sup>12</sup> Giovanni Pitari,<sup>13</sup> Helen Rogers,<sup>14</sup> Matthew Johnson,<sup>14</sup> John Pyle,<sup>14</sup> Kathy Law,<sup>14</sup> Michiel van Weele,<sup>15</sup> and Oliver Wild<sup>16</sup>

Received 15 September 2002; revised 3 December 2002; accepted 17 December 2002; published 31 January 2003.

[1] Ozone is an air quality problem today for much of the world's population. Regions can exceed the ozone air quality standards (AQS) through a combination of local emissions, meteorology favoring pollution episodes, and the clean-air baseline levels of ozone upon which pollution builds. The IPCC 2001 assessment studied a range of global emission scenarios and found that all but one projects increases in global tropospheric ozone during the 21st century. By 2030, near-surface increases over much of the northern hemisphere are estimated to be about 5 ppb (+2 to +7 ppb over the range of scenarios). By 2100 the two more extreme scenarios project baseline ozone increases of >20 ppb, while the other four scenarios give changes of -4 to +10 ppb. Even modest increases in the background abundance of tropospheric ozone might defeat current AQS strategies. The larger increases, however, would gravely threaten both urban and rural air quality over most of the northern hemisphere. **INDEX TERMS:** 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 1610 Global Change: Atmosphere (0315, 0325). **Citation:** Prather, M., et al., Fresh air in the 21st century?, *Geophys. Res. Lett.*, 30(2), 1100, doi:10.1029/2002GL016285, 2003.

### 1. Introduction

[2] The air we breathe can contain noxious substances in the form of trace gases and aerosols. Ozone (O<sub>3</sub>) is

identified as one of the more serious of these air pollutants, and the large abundances of O<sub>3</sub> observed within and downwind of metropolitan regions are clearly identified with emissions of ozone precursors, specifically, oxides of nitrogen (NO<sub>x</sub>), carbon monoxide (CO) and volatile organic compounds (VOC), by the industrial and transportation sectors [*Haagen-Smit*, 1951]. Breathing ozone, even at relatively low abundances, is correlated with pulmonary damage and asthma attacks [e.g., *Peden*, 2001; *Desqueyroux et al.*, 2002; *Mortimer et al.*, 2002]. Ground-level O<sub>3</sub> damages agricultural crops and natural ecosystems [e.g., *Mauzerall and Wang*, 2001; *Oksanen and Holopainen*, 2001] and reacts with environmental compounds to produce other toxic substances [e.g., *Morrison and Nazaroff*, 2002].

[3] Ozone is an air quality problem today for much of the world's population. For example, in the U.S. the 8-hour ozone standard of 80 ppb (all abundances are mole fraction, ppb = 10<sup>-9</sup>) was exceeded over the last decade by an average 28 days per year for New England, where "serious" to "severe" non-attainment areas cover the populous regions [U.S. Environmental Protection Agency, Region 1: New England, <http://www.epa.gov/region01/eco/ozone/>]. In Europe, more than half the urban population is exposed to ozone above the 8-hour standard of 55 ppb for more than 30 days per year [*EEA*, 2002]. Some developing countries like India have air quality standards (AQS) for SO<sub>2</sub>, NO<sub>2</sub>, aerosols, but not O<sub>3</sub>. The impacts of O<sub>3</sub> pollution on vegetation and in developing countries is only recently being assessed [e.g., *Emberson et al.*, 2001; *Taylor*, 2001]. The global damages caused by increasing O<sub>3</sub> levels have not been fully evaluated.

[4] Strategies to abate urban O<sub>3</sub> originally focused on local or regional solutions such as the State of California's Air Quality Management Districts (<http://www.aqmd.gov/>). The 1979 Geneva Convention on Long-Range Transboundary Air Pollution (<http://www.unece.org/env/lrtap/>) now has Protocols that consider continental-scale transport of O<sub>3</sub> and its precursors. Recent studies have shown that transoceanic, intercontinental transport of O<sub>3</sub> and related pollutants couples the major continents of the northern mid-latitudes [*Tarrason and Iversen*, 1998; *Wild and Akimoto*, 2001; *Li et al.*, 2002] and hint that the O<sub>3</sub> AQS may be a global problem. Still, the typical scientific study to aid policy decisions on future O<sub>3</sub> AQS [e.g., *Jensen et al.*, 2001] does not consider global-scale changes in tropospheric O<sub>3</sub> that are anticipated during the 21st century.

[5] More than a dozen research groups analyzed the changes in atmospheric chemistry projected for the 21st century as part of the OxComp workshop of the Third Assessment Report of the Intergovernmental Panel on

<sup>1</sup>Department of Earth System Science, UC Irvine, California, USA.

<sup>2</sup>Institutt for Geofysikk, University of Oslo, Oslo, Norway.

<sup>3</sup>Swiss Federal Institute of Technology, Lausanne, Switzerland.

<sup>4</sup>Max Planck Institute for Meteorology, Hamburg, Germany.

<sup>5</sup>Joint Research Centre, Environment Institute, Ispra, Italy.

<sup>6</sup>The Hadley Centre, UK Met Office, Bracknell, United Kingdom.

<sup>7</sup>Now at Inst. für Meteorologie, Free Univ., Berlin, Germany.

<sup>8</sup>Institut Pierre Simon Laplace, CEA-CNRS, Gif-sur-Yvette, France.

<sup>9</sup>Geophysical Fluid Dynamics Laboratory, NOAA, Princeton, New Jersey, USA.

<sup>10</sup>Dept of Earth & Planetary Sciences, Harvard U., Cambridge, Maryland, USA.

<sup>11</sup>Max Planck Institute for Chemistry, Mainz, Germany.

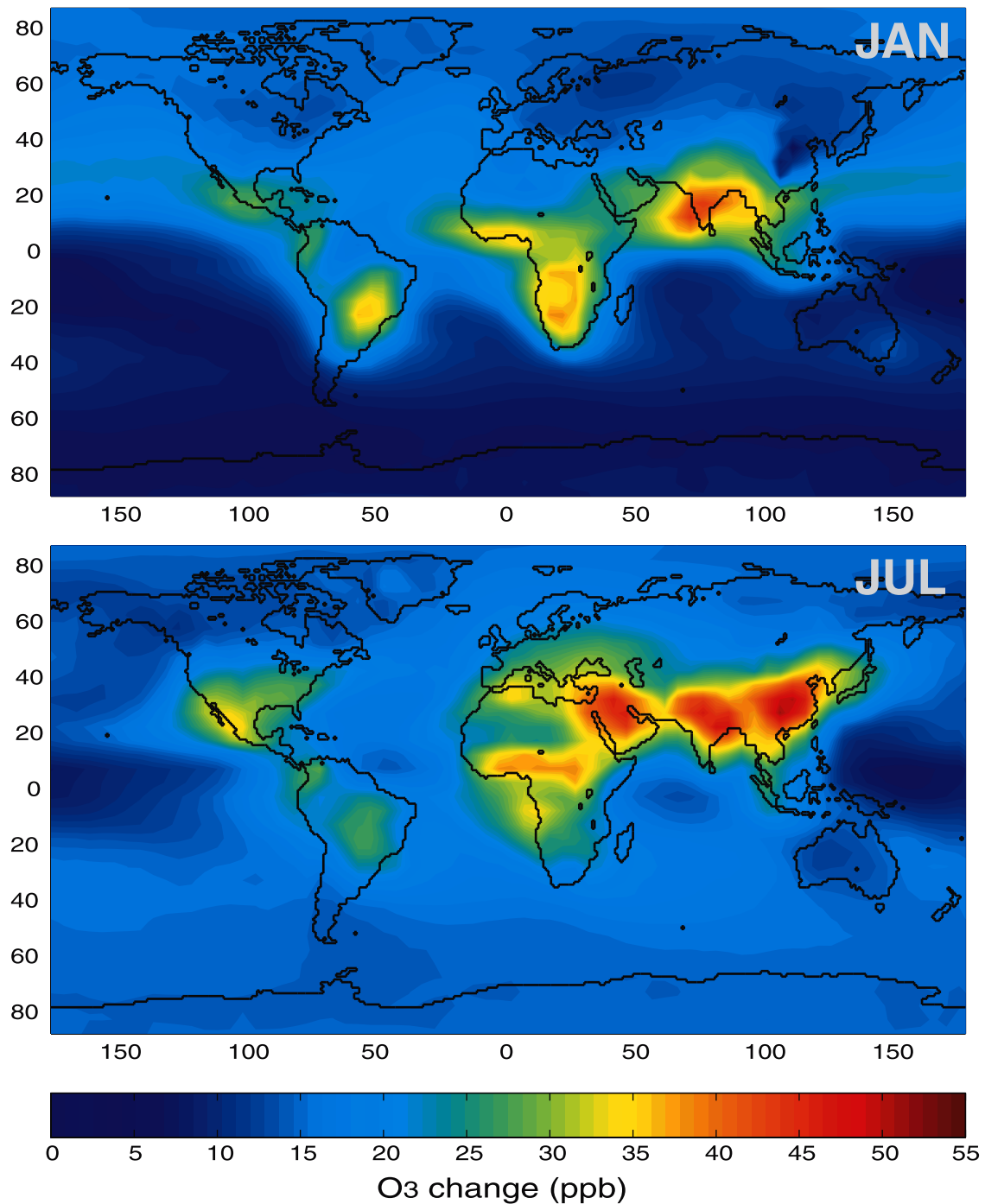
<sup>12</sup>Belgian Institute for Space Aeronomy, Brussels, Belgium.

<sup>13</sup>Dipartimento di Fisica, U.L'Aquila, Coppito, L'Aquila, Italy.

<sup>14</sup>Centre for Atmos. Sci., Cambridge U., Cambridge, United Kingdom.

<sup>15</sup>Royal Netherlands Meteorological Inst., De Bilt, The Netherlands.

<sup>16</sup>Frontier Research System for Global Change, Yokohama, Japan.



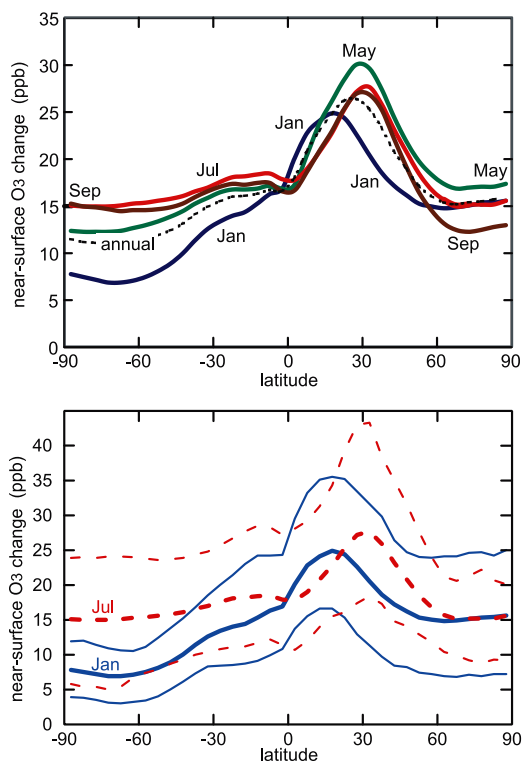
**Figure 1.** Monthly mean surface O<sub>3</sub> increase (ppb) for Jan and Jul from Y2000 to Y2100 following scenario A2x. Results are the average of 10 models [Prather and Ehhalt, 2001]: HGIS, IASB, KNMI, MOZ1, MOZ2, UCAM, UCI, UIO1, UKMO, ULAQ.

Climate Change (IPCC/TAR) [Prather and Ehhalt, 2001]. Three-dimensional global models were used to project the changes in greenhouse gases resulting from changing anthropogenic emissions based on the Special Report on Emissions Scenarios (SRES) [Nakićenović *et al.*, 2000]. The IPCC/TAR reported tropospheric O<sub>3</sub> column as a climate forcing and only noted briefly that surface O<sub>3</sub> increases by 2100 might be “threatening attainment of air quality standards over most metropolitan and even rural regions...”

Surface maps and seasonality of these projected O<sub>3</sub> changes are presented and discussed here for the first time.

## 2. Results

[6] The increases in near-surface O<sub>3</sub> projected from year 2000 to year 2100 for one of the extreme SRES scenarios are shown in Figure 1 as January and July monthly averages. The largest increases typically occur in the tropics



**Figure 2.** (top) Zonal-mean surface  $O_3$  increase (ppb) from Y2000 to Y2100 as a function of latitude following scenario A2x. See Figure 1. The 4 months shown (Jan(blue), May(green), Jul(red), Sep(brown)) bound all other months and are labeled at their extrema. The annual average (dashed) is also shown. (bottom) The Jan zonal average (blue, thick solid) and Jul zonal average (red, thick dashed) are shown again with their minima and maxima (thin lines). These min/max are the zonal averages of the extreme model result for each  $5^\circ \times 5^\circ$  grid box.

where high sunlight conditions prevail, but the rise in  $O_3$  affects most of the globe, including what are currently thought of as clean-air remote regions. These values are taken from global tropospheric chemistry model simulations for the IPCC/TAR scenario A2x, which was the only scenario analyzed with the full chemistry models [Prather and Ehhalt, 2001]. This projection of anthropogenic emissions is based on the preliminary SRES A2 scenario and a preliminary estimate of the methane ( $CH_4$ ) abundance. Prior to the completion of the TAR the SRES scenarios were revised and the  $CH_4$  abundances were recalculated to include the emissions of  $NO_x$ , CO, and VOC.

[7] In all, fourteen independent models contributed results to the IPCC/TAR calculation of tropospheric  $O_3$  change using the SRES emission scenarios. The meteorology and spatial resolution of these models range widely, they include both assimilated and climate-model winds, and most resolve the meteorology at 6 hours or better. Of these, ten (HGIS, IASB, KNMI, MOZ1, MOZ2, UCAM, UCI, UIO1, UKMO, ULAQ) gave complete calculations and diagnostics for scenario A2x, and their average is analyzed here. Results for the contemporary atmosphere (year 2000) were quite realistic in simulating observations of  $O_3$  and CO at remote sites. While results vary from model to model, the basic pattern and magnitude of tropospheric  $O_3$  changes in

these ten models is similar (see figures 4.12–13 of Prather and Ehhalt [2001]), and thus the average is a robust estimate for these emissions. The differing treatment and resolution of chemistry in the boundary layer of these models prevented a meaningful comparison of ground-level  $O_3$ . Instead, we reported the average over 0 to 2 km in altitude and expect this abundance to be representative of the time-mean ground-level  $O_3$  except over intense emission regions. These averages, at  $5^\circ$  latitude by  $5^\circ$  longitude resolution, are available along with the minimum and maximum range of the ten models.

[8] The tropical hot spots in Figure 1 can be explained by the shifting pattern of emissions of  $NO_x$  and CO in the SRES emission patterns for scenario A2x. Anthropogenic  $NO_x$  emissions increase 3.4-fold from year 2000 to year 2100. Relative increases in Europe (2.0) and North America (1.8) are less than the average, with relative increases in India (5.9), Southeast Asia (5.7), Africa (7.7), and South America (8.7) driving the total. Anthropogenic CO emissions increase 2.4-fold from year 2000 to year 2100. Relative increases in Europe (2.0) and North America (1.6) are likewise below the average; but relative increases in India (4.4), Southeast Asia (4.6), Africa (6.0), and South America (6.5) are much greater. These scenarios project an overall shift in precursors to lower latitudes with consequent impact on  $O_3$  as noted previously [Gupta et al., 1998].

[9] The zonal mean increases in near-surface  $O_3$  are shown in Figure 2 (top) for a selection of months that bound the annual range. For the summer months (May–Sep) from  $25^\circ N$  to  $40^\circ N$  the increase is larger than 25 ppb, with even greater increases in the vicinity of continents. Even in remote southern latitudes poleward of  $50^\circ S$  the increase ranges from 7 to 16 ppb. Figure 2 (bottom) shows the extreme ranges of all ten model results for Jan (dashed) and Jul (solid), selecting the extreme of all models for each  $5^\circ \times 5^\circ$  grid box to average zonally. Thus, based on the range of independent model results, the 2/3-likelihood confidence interval for the zonal mean increase in near-surface  $O_3$  due to these emissions is judged to be less than  $\pm 5$  ppb. As identified in the OxComp study, the increase in  $CH_4$  abundance from 1750 ppb to 4300 ppb drives almost half of this  $O_3$  increase, and it is primarily the increased emissions of  $NO_x$  rather than CO or VOC that drive the other half.

### 3. Discussion

[10] What do projections of global tropospheric  $O_3$  mean for AQS in the 21st century? Clearly, these increases are not intended as direct projections of urban  $O_3$  change, but rather, they indicate an upward shift in the baseline levels of  $O_3$  upon which regional pollution builds. The extreme, high- $O_3$  events as measured in the hourly AQS are driven by local emissions and less likely affected by baseline  $O_3$  increases. Efforts to meet the new 8-hour standards, however, and particularly the European cumulative standards for crops and vegetation (AOT40), would be greatly impacted by baseline increases of 15 to 25 ppb. It is worrisome that the largest baseline increases occur in northern mid-latitudes around continents during summer when air quality is currently at its worst. There are already agricultural concerns about the summertime high  $O_3$  in developing countries [Mauzerall and Wang, 2001]. In addition, the high  $O_3$



regions coincide with increasing aerosol emissions [Penner, 2001], which further exacerbates human health impacts.

[11] How representative is this high-end scenario of future O<sub>3</sub> increases? Among the six final SRES illustrative scenarios, two (A2 and A1FI) have emissions of O<sub>3</sub> precursors as large as the A2x scenario shown here. If we scale the change in near-surface O<sub>3</sub> with that of the total tropospheric O<sub>3</sub> as reported in the TAR [Prather and Ehhalt, 2001], then these two scenarios would project changes like those illustrated here for the latter decades of the 21st century. On the other hand, the four less extreme SRES scenarios (A1B, A1T, B1, B2) project much lower fossil fuel use and smaller near-surface O<sub>3</sub> changes ranging from -4 to +10 ppb by 2100. More immediately, by 2030 all six scenarios would project near-surface O<sub>3</sub> increases that range from 9% to 34% (averaging 25%) of those shown here. Such near-term increases of about 5 (+2 to +7) ppb over the northern hemisphere would notably impede AQS attainment [Fiore et al., 2002]. One critical element in these projections is the scenario for emissions of O<sub>3</sub> precursors, not just their amount, but their location. A known flaw with the SRES scenarios is that they did not explicitly consider the emergence of new ozone-related air quality regulations in developing countries. A revised and expanded SRES-like effort, one that focuses on the regional emissions of O<sub>3</sub> precursors and allows for emerging air quality regulations, is required for AQS planning over the next several decades.

[12] What is missing? These projections of 21st century changes in global tropospheric chemistry include only changes in anthropogenic emissions. As noted in the TAR, the response of the climate system to the overall anthropogenic forcing (including carbon dioxide) is expected to be larger than anything observed in the last millennium, and thus we expect natural ecosystems and their emissions of O<sub>3</sub> precursors to be altered. Unfortunately, we were unable to evaluate this feedback in the TAR due to a lack of research and publication on this topic. In addition, the physical climate change itself will alter the dynamics, temperature, and humidity of the troposphere, including possibly the occurrence of stagnation episodes that lead to AQS exceedences. One of the OxComp models (UKMO) has shown since the TAR that the 21st century physical climate change driven by the A2 scenario tends to reduce global tropospheric O<sub>3</sub> because of the higher humidity and temperature [Johnson et al., 2001], a result consistent with earlier sensitivity studies [Brasseur et al., 1998]. These missing feedbacks represent a major source of uncertainty in projecting near-surface baseline O<sub>3</sub> increases. They need to be evaluated within the research of the broader community before the next assessments.

[13] **Acknowledgments.** The authors thank the leadership of Sir John Houghton in Working Group I of the IPCC Third Assessment Report, which made these collaborative model studies possible.

## References

- Brasseur, G. P., et al., Past and future changes in global tropospheric ozone: Impact on radiative forcing, *Geophys. Res. Lett.*, 25, 3807–3810, 1998.
- Desqueyroux, H., J. C. Pujet, M. Prosper, F. Squinazi, and I. Momas, Short-term effects of low-level air pollution on respiratory health of adults suffering from moderate to severe asthma, *Environ. Res.*, 89, 29–37, 2002.
- EEA, Environmental signals 2002: Benchmarking the millennium, Chapter 10, Air Pollution, No. 9 Environmental Assessment Report, pp. 73–84, <http://europa.eu.int>, European Environment Agency, Copenhagen, Denmark, 2002.
- Emberson, L. D., et al., Impacts of air pollutants on vegetation in developing countries, *Water & Soil Pollut.*, 130, 107–118, 2001.
- Fiore, A. M., et al., Linking ozone air pollution and climate change: The case for controlling methane, *Geophys. Res.*, 29, 10.1029/2002GL015601, in press, 2002.
- Gupta, M. L., R. J. Cicerone, and S. Elliott, Perturbation to global tropospheric oxidizing capacity due to latitudinal redistribution of surface sources of NO<sub>x</sub>, CH<sub>4</sub> and CO, *Geophys. Res. Lett.*, 28, 3931–3934, 1998.
- Haagen-Smit, A. J., Chemistry and physiology of Los Angeles smog, *Indust. Eng. Chem.*, 44, 1342–1346, 1951.
- Jensen, S. S., R. Berkowicz, M. Winther, F. Palmgren, and Z. Zlatev, Future air quality in Danish cities due to new emission and fuel quality directives of the European Union, *International J. Vehicle Design*, 27, 195–208, 2001.
- Johnson, C. E., D. S. Stevenson, W. J. Collins, and R. G. Derwent, Role of climate feedback on methane and ozone studied with a coupled ocean-atmosphere-chemistry model, *Geophys. Res. Lett.*, 28, 1723–1726, 2001.
- Li, Q., et al., Transatlantic transport of pollution and its effects on surface ozone in Europe and North America, *J. Geophys.*, 107, 10.1029/2001JD001422, 2002.
- Mauzerall, D. L., and X. P. Wang, Protecting agricultural crops from the effects of tropospheric ozone exposure: Reconciling science and standard setting in the United States, Europe, and Asia, *Ann. Rev. Energy & Environ.*, 26, 237–268, 2001.
- Morrison, G. C., and W. W. Nazaroff, Ozone interactions with carpet: Secondary emissions of aldehydes, *Env. Sci. & Tech.*, 36, 2185–2192, 2002.
- Mortimer, K. M., L. M. Neas, D. W. Dockery, S. Redline, and I. B. Tager, The effect of air pollution on inner-city children with asthma, *Eur. Respiratory J.*, 19, 699–705, 2002.
- Nakićenović, N., et al., Special Report on Emissions Scenarios, Cambridge U. Press, Cambridge, 2000.
- Oksanen, E., and T. Holopainen, Responses of two birch (*Betula pendula* Roth) clones to different ozone profiles with similar AOT40 exposure, *Atm. Environ.*, 35, 5245–5254, 2001.
- Peden, D. B., Air pollution in asthma: effect of pollutants on airway inflammation, *Annals Allergy Asthma & Immunology*, 87, 12–17, 2001.
- Penner, J. E., Chapter 5. Aerosols, their Direct and Indirect Effects, in "Climate Change 2001: The Scientific Basis", pp. 289–348, edited by J. T. Houghton et al., Cambridge U. Press, Cambridge, 2001.
- Prather, M., and D. Ehhalt, Chapter 4. Atmospheric Chemistry and Greenhouse Gases, in "Climate Change 2001: The Scientific Basis", pp. 239–287, edited by J. T. Houghton et al., Cambridge U. Press, Cambridge, 2001.
- Tarrason, L., and T. Iversen, Modelling intercontinental transport of atmospheric sulphur in the northern hemisphere, *Tellus B*, 50, 331–352, 1998.
- Taylor, G. E., Risk assessment of tropospheric ozone: Human health, natural resources, and ecology, *Human & Ecol. Risk Assess.*, 7, 1183–1193, 2001.
- Wild, O., and H. Akimoto, Intercontinental transport of ozone and its precursors in a three-dimensional global CTM, *J. Geophys. Res.*, 106, 27,729–27,744, 2001.

M. Prather, Dept of Earth System Science, UC Irvine, CA 92697, USA.  
M. Gauss, T. Berntsen, I. Isaksen, and J. Sundet, Inst. for Geofysikk, U. of Oslo, 0315, Oslo, Norway.

I. Bey, Swiss Federal Inst. of Technology, Lausanne, Switzerland.

G. Brasseur, Max Planck Institute for Meteorology, 20146, Hamburg, Germany.

F. Dentener, Joint Research Centre, Environment Institute, I-21020, Ispra, Italy.

R. Derwent and D. Stevenson, The Hadley Centre, UK Met Office, Bracknell, UK.

L. Grenfell, Inst. für Meteorologie, Free University, 12165, Berlin, Germany.

D. Hauglustaine, Institut Pierre Simon Laplace, LSCE, CEA-CNRS, Gif-sur-Yvette, France.

L. Horowitz, Geophys. Fluid Dynamics Lab., NOAA, Princeton, NJ, USA.

D. Jacob and L. Mickley, Dept. of Earth and Planetary Sciences, Harvard U., Cambridge, MA, USA.

M. Lawrence and R. von Kuhlmann, Max Planck Inst. for Chemistry, Mainz, Germany.

J.-F. Muller, Belgian Inst. for Space Aeronomy, B-1180, Brussels, Belgium.

G. Pitari, Dipartimento di Fisica, U. L'Aquila, 67010, Coppito, L'Aquila, Italy.

H. Rogers, M. Johnson, J. Pyle, and K. Law, Center for Atmospheric Science, Cambridge U., Cambridge CB2 1EW, UK.

M. van Weele, Royal Netherlands Meteorological Institute (KNMI), 3730, AE De Bilt, The Netherlands.

O. Wild, Frontier Research System for Global Change, Yokohama, Japan.