

## Multimodel ensemble simulations of present-day and near-future tropospheric ozone

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[1] Global tropospheric ozone distributions, budgets, and radiative forcings from an ensemble of 26 state-of-the-art atmospheric chemistry models have been intercompared and synthesized as part of a wider study into both the air quality and climate roles of ozone. Results from three 2030 emissions scenarios, broadly representing “optimistic,” “likely,” and “pessimistic” options, are compared to a base year 2000 simulation. This base case realistically represents the current global distribution of tropospheric ozone. A further set of simulations considers the influence of climate change over the same time period by forcing the central emissions scenario with a surface warming of around 0.7K. The use of a large multimodel ensemble allows us to identify key areas of uncertainty and improves the robustness of the results. Ensemble mean changes in tropospheric ozone burden between 2000 and 2030 for the 3 scenarios range from a 5% decrease, through a 6% increase, to a 15% increase. The intermodel uncertainty ( $\pm 1$  standard deviation) associated with these values is about  $\pm 25\%$ . Model outliers have no significant influence on the ensemble mean results. Combining ozone and methane changes, the three scenarios produce radiative forcings of  $-50$ ,  $180$ , and  $300 \text{ mW m}^{-2}$ , compared to a  $\text{CO}_2$  forcing over the same time period of  $800\text{--}1100 \text{ mW m}^{-2}$ . These values indicate the importance of air pollution emissions in short- to medium-term climate forcing and the potential for stringent/lax control measures to improve/worsen future climate forcing. The model sensitivity of ozone to imposed climate change varies between models but modulates zonal mean mixing ratios by  $\pm 5$  ppbv via a variety of feedback mechanisms, in particular those involving water vapor and stratosphere-troposphere exchange. This level of climate change also reduces the methane lifetime by around 4%. The ensemble mean year 2000 tropospheric ozone budget indicates chemical production, chemical destruction, dry

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deposition and stratospheric input fluxes of 5100, 4650, 1000, and 550 Tg(O<sub>3</sub>) yr<sup>-1</sup>, respectively. These values are significantly different to the mean budget documented by the Intergovernmental Panel on Climate Change (IPCC) Third Assessment Report (TAR). The mean ozone burden (340 Tg(O<sub>3</sub>)) is 10% larger than the IPCC TAR estimate, while the mean ozone lifetime (22 days) is 10% shorter. Results from individual models show a correlation between ozone burden and lifetime, and each model's ozone burden and lifetime respond in similar ways across the emissions scenarios. The response to climate change is much less consistent. Models show more variability in the tropics compared to midlatitudes. Some of the most uncertain areas of the models include treatments of deep tropical convection, including lightning NO<sub>x</sub> production; isoprene emissions from vegetation and isoprene's degradation chemistry; stratosphere-troposphere exchange; biomass burning; and water vapor concentrations.

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## 1. Introduction

[2] The global environmental issues of climate change and air pollution are closely linked, not least by the trace gas ozone (O<sub>3</sub>). Ozone concentrations in the troposphere are thought to have increased significantly since preindustrial times, as a direct result of human activities [Volz and Kley, 1988; Staehelin et al., 1994; Lamarque et al., 2005a]. Tropospheric O<sub>3</sub> increases represent the third largest greenhouse gas contribution to radiative forcing of climate change over this time period, with a forcing equivalent to about 24% of that from carbon dioxide [Ramaswamy et al., 2001]. Ozone is also a serious and ubiquitous air pollutant, affecting the respiratory health of a large proportion of the world population [World Health Organization, 2003], reducing the yields of staple agricultural crops such as wheat and soy bean, and damaging natural ecosystems [Emberson et al., 2003; Wang and Mauzerall, 2004].

[3] There are two sources of tropospheric ozone: transport from the stratosphere, and in situ chemical production. Ozone production takes place when carbon monoxide (CO) and hydrocarbons are photo-oxidized in the presence of nitrogen oxides (NO<sub>x</sub> = NO + NO<sub>2</sub>) [Crutzen, 1974; Liu et al., 1980; Atkinson, 2000]. The ozone budget is closed by two loss processes: dry deposition to the Earth's surface, and chemical destruction. Ozone destruction occurs mainly via reactions with water vapor (following photolysis) and with hydrogen peroxy and hydroxyl radicals (HO<sub>x</sub> = HO<sub>2</sub> + OH). Integrated over the whole troposphere, chemical production and loss rates are several times larger than the influx from the stratosphere and the surface deposition flux.

[4] The main ozone precursors (CO, hydrocarbons, and NO<sub>x</sub>) are all emitted as by-products of human activities, as well as having significant natural sources. The same anthropogenic and natural emissions also influence HO<sub>x</sub>, affecting ozone destruction. The rapid growth in anthropogenic emissions following industrialization has been the major driver of increases in tropospheric ozone, and has been the subject of intensive study [e.g., Levy et al., 1985; Crutzen and Zimmerman, 1991; Lelieveld and Dentener, 2000; Lamarque et al., 2005a]. Interest has more recently turned to the potential influence of climate change on future levels of ozone [Johnson et al., 1999, 2001; Isaksen et al., 2003; Grenfell et al., 2003; Stevenson et al., 2005; Hauglustaine et al., 2005; Murazaki and Hess, 2006].

Warming will increase water vapor concentrations, and changes in temperature and water vapor will affect the reaction rates of many chemical conversions. Climate change may also alter global circulation dynamics, changing several processes that govern the distribution of tropospheric ozone, such as stratosphere-troposphere exchange, the distribution of convection, and ventilation of the boundary layer. Changes in climate will also affect many of the natural sources of trace gases, such as wetland CH<sub>4</sub> [Walter and Heimann, 2000; Gedney et al., 2004; Shindell et al., 2004], biogenic volatile organic compounds [Sanderson et al., 2003; Wiedinmyer et al., 2005; Lathière et al., 2005], lightning NO<sub>x</sub> [Price et al., 1997] and soil NO<sub>x</sub> [Ganzeveld et al., 2002].

[5] Global atmospheric chemistry models currently provide our best estimates of tropospheric distributions of trace species such as ozone and the hydroxyl radical (OH). These models underpin our understanding of the oxidizing capacity of the atmosphere, and radiative forcings arising from changes to methane (CH<sub>4</sub>) and O<sub>3</sub> concentrations [e.g., Prather et al., 2001, 2003; Gauss et al., 2003]. As such, they constitute the best tools at our disposal for making quantitative future projections of tropospheric composition.

[6] Such models are only approximations of the real atmosphere, and exhibit uncertainties arising from many processes, including emissions, transport (resolved advection and subgrid-scale convection), chemistry (photochemical, gas phase, aqueous phase, and heterogeneous reactions), mixing, deposition, and also from upper and lower boundary conditions. All these processes are interrelated, and tend to be handled by specific models in different ways. Consequently, models show a range of sensitivities to changes in, for example, future trace gas emissions and climate.

[7] Some previous model intercomparison studies have focused on comparisons of models and measurements [e.g., Brunner et al., 2003, 2005; Roelofs et al., 2003; Kinne et al., 2005; Textor et al., 2005], but this is not the main thrust of the work presented here. Instead, this paper intercompares and synthesizes simulated ozone results from a wide cross section of state-of-the-art models, and is part of a larger study [Dentener et al., 2006a] coordinated by the European Union project Atmospheric Composition Change: the European Network of excellence (ACCENT; <http://www.accent-network.org>). The use of a wide range of

differently formulated models allows us to increase the robustness of future projections, and quantify levels of uncertainty. Other aspects of this wider modeling study include a detailed analysis of surface ozone, including impacts on human health and vegetation (K. Ellingsen et al., Ozone air quality in 2030: A multi-model assessment of risks for health and vegetation, manuscript in preparation, 2006); a comparison of modeled NO<sub>2</sub> tropospheric columns with satellite observations [van Noije et al., 2006]; a comparison of modeled and measured CO (D. Shindell et al., Multi-model simulations of carbon monoxide: Comparison with observations and projected near-future changes, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Shindell et al., submitted manuscript, 2006); and an analysis of simulated deposition budgets of NO<sub>y</sub>, SO<sub>x</sub>, and NH<sub>x</sub> (F. Dentener et al., Nitrogen and sulfur deposition on regional and global scales: A multimodel evaluation, submitted to *Global Biogeochemical Cycles*, 2005, hereinafter referred to as Dentener et al., submitted manuscript, 2005). A parallel study [Gauss et al., 2005] intercompares ozone from a similar range of models from preindustrial to present day, with a particular focus on both tropospheric and stratospheric changes.

[8] In the following section, we briefly describe some of the pertinent features of the participating models, emphasizing potentially important differences in their formulation. In section 3, we describe the model simulations that were carried out, before reporting and discussing results in section 4. Section 4 firstly describes how we have processed results from a diverse range of models, and created ensemble means and standard deviations. Ozone results for the year 2000 are then described and compared to ozonesonde observations. We then consider results from the 2030 scenarios, in terms of changes in both emissions and climate relative to 2000. We present tropospheric ozone budgets and methane lifetimes, and calculate radiative forcings for the various cases. The last section presents a summary and conclusions.

## 2. Participating Models: Sources of Differences

[9] Results from a total of 26 different models (see Appendix A and Table A1) are analyzed here. These 26 models include some cases where the same “core” model was used in two or more different configurations; e.g., the models CHASER, GMI, LMDz/INCA and MATCH-MPIC were driven by a variety of different underlying meteorological fields. Nearly all the models were set up as chemistry-transport models (CTMs); in these models the meteorology is not influenced by the chemical fields. Only two models (CHASER\_GCM and ULAQ) were set up as fully coupled chemistry-climate models (CCMs), where the chemical fields enter the radiation calculations of the driving general circulation model (GCM), and directly influence the dynamics. For ULAQ, this fully coupled mode was only switched on for the climate change simulation (S5; simulations are described in section 3). Of the CTMs, thirteen were driven by numerical weather prediction (NWP) data, with nine using European Centre for Medium-Range Weather Forecasts (ECMWF) analyses or forecasts. The other thirteen models were driven by GCM output, with eleven different driving GCMs. Of these models, ten

simulated the climate change scenario. When available, each of the 26 model results is given equal weight in the following analysis. We also check for the potential influence of model outliers on our conclusions.

[10] Global horizontal resolution ranged from 10° × 22.5° (ULAQ) to 1.9° × 1.9° (MOZECH, MOZ2-GFDL), with one model (TM5) operating a 1° × 1° nested grid over Europe, North America and Asia. The number of vertical levels ranged from 9 (STOCHEM\_HadAM3) to 60 (MATCH models), with typically 20 levels of variable thickness in the troposphere, increasing in resolution toward the surface. The lowest layer depth ranged from about 35 to 800 m. Studies have shown that, all other things being equal, higher-resolution models tend to produce less ozone from the same levels of precursor emissions, due to the lower levels of forced “mixing” generated when emissions are added to large grid boxes [e.g., Liang and Jacobsen, 2000; Esler, 2003]. We might therefore expect the lower-resolution models to be inherently more mixed and hence more chemically active than the higher-resolution models.

[11] The cross-tropopause flux of ozone [Gettelman et al., 1997; Roelofs et al., 2003; Olsen et al., 2004] is a key determinant of upper tropospheric ozone concentrations. Models used a wide variety of upper boundary conditions. Two models (LLNL-IMPACT and ULAQ) included detailed stratospheric chemistry. The UIO\_CTM2 model used stratospheric profiles from an earlier run with full stratospheric chemistry. The FRSGC/UCI model used the “linearized ozone chemistry” (LINOZ) scheme [McLinden et al., 2000], which can be considered a simplified stratospheric chemistry scheme. Some models employed “prescribed flux” methods: GEOS-CHEM and GMI used the “synthetic ozone” (SYNOZ) scheme [McLinden et al., 2000]. STOCHEM-HadAM3 added an ozone flux at 100 hPa, calculated from an ozone climatology and the local vertical winds. Other models fixed ozone concentrations above a certain level: p-TOMCAT (<10 hPa); UM\_CAM, MATCH-MPIC (<30 hPa); IASB (<50 hPa); and GISS (above tropopause). Another method used was to relax or nudge toward a climatology above a certain level: CHASER, TM5 (<50 hPa); LMDz/INCA (>380 K isentrope; <~100 hPa); TM4 (<60 hPa (30°S–30°N); <123 hPa elsewhere); and STOCHEM\_HadGEM (above tropopause). Finally, some models used a combination of fixed ozone above a certain level and relaxation below this level: MOZ2-GFDL (fixed <14 hPa, relax between 14 hPa and tropopause), MOZECH and MOZECH2 (fixed <30 hPa, relax <100 hPa in the tropics and <200 hPa in the extratropics), MOZART4 (fixed <50 hPa, relax between 50 hPa and tropopause). A relaxation e-folding timescale of 3–10 days is typically applied. The relative merits of these different schemes are not well known, but some of these variants have been compared in the consistent framework of ERA-40 (45-year ECMWF reanalysis [Simmons and Gibson, 2000]) by van Noije et al. [2004].

[12] Deposition of ozone to the Earth’s surface and biosphere strongly influences boundary layer ozone concentrations. Models used deposition schemes of varying levels of sophistication, but all used “resistance” type formulations [e.g., Wesely, 1989] coupled to prescribed land cover distributions. Inspection of ozone deposition distri-



**Table 1.** Specifications of the Simulations<sup>a</sup>

Code	Name	Meteorology	Emissions	Reference
S1	Y2000	2000/1995–2004	2000 (EDGAR3.2)	<i>Olivier and Berdowski</i> [2001]
S2	CLE	2000/1995–2004	2030 IIASA CLE	<i>Dentener et al.</i> [2005]
S3	MFR	2000/1995–2004	2030 IIASA MFR	<i>Dentener et al.</i> [2005]
S4	A2	2000/1995–2004	2030 SRES A2	<i>Nakicenovic et al.</i> [2000]
S5	CLE-ΔC	2025–2034	2030 IIASA CLE	<i>Stevenson et al.</i> [2005]

<sup>a</sup>The code and the name are used interchangeably in the text.

butions from the models suggest that the schemes generate quite variable deposition velocities over different terrains, although global deposition fluxes are relatively consistent (see budget analysis below).

[13] Convective mixing of ozone and its precursors exerts a major control on the vertical distribution of tropospheric ozone [*Lelieveld and Crutzen*, 1994; *Lawrence et al.*, 2003; *Olivie et al.*, 2004; *Doherty et al.*, 2005]. All models include parametrizations for convection; the most commonly used (by seven of the models) is the mass flux scheme of *Tiedtke* [1989], sometimes with modifications. Some model convection schemes are tuned using vertical profiles of the short-lived isotope <sup>222</sup>Rn [e.g., *Jacob et al.*, 1997; *Rasch et al.*, 2000]; however, observations of <sup>222</sup>Rn are sparse, and soil emissions are not well known [e.g., *Robertson et al.*, 2005]. In addition, convection over oceanic areas is essentially not considered, as the <sup>222</sup>Rn source is land-based. Consequently, convection schemes are poorly constrained, and are probably a major source of differences between models. Model uncertainties associated with convection may be exacerbated because convection is often colocated with lightning and tropical vegetation, key natural sources of the ozone precursors NO<sub>x</sub> and isoprene (C<sub>5</sub>H<sub>8</sub>). Convective mixing promotes interaction between these two sources throughout the depth of the tropical troposphere, with potentially important implications for ozone [e.g., *von Kuhlmann et al.*, 2004; *Doherty et al.*, 2005]. Model uncertainties associated with convection also include wash-out of key species (e.g., nitric acid, HNO<sub>3</sub>, and hydrogen peroxide, H<sub>2</sub>O<sub>2</sub>) [e.g., *Sudo and Takahashi*, 2001].

[14] Chemical schemes continue to develop in complexity through the addition and expansion of nonmethane hydrocarbon (NMHC) schemes [e.g., *Houweling et al.*, 1998; *Wang et al.*, 1998; *Collins et al.*, 1999; *Roelofs and Lelieveld*, 2000; *von Kuhlmann et al.*, 2003a], and the inclusion of aerosol modules, allowing gas-aerosol interactions [e.g., *Tie et al.*, 2001, 2003; *Martin et al.*, 2003; *Bauer et al.*, 2004; *Bell et al.*, 2005]. All the models include some description of NMHC chemistry, but they range from relatively highly parameterized “lumped” schemes (e.g., GISS: 35 species) to more detailed schemes (e.g., MOZART4: 96 species). Isoprene degradation schemes in the models show a similar range of complexity, and this may account for some variations in tropical ozone [e.g., *Pöschl et al.*, 2000; *von Kuhlmann et al.*, 2004]. Photolysis schemes within models are also parametrized in a variety of ways, handling absorption and scattering of radiation by clouds and aerosols differently. About half of the models include some aerosol chemistry (Table A1). It is unclear how important the differences in the current models’ chemical schemes are in generating differences in the distributions of key species such as O<sub>3</sub> and OH, and how the schemes contribute to differing sensitivities to emissions

and climate change. Intercomparisons of specific chemical modules [e.g., *Olson et al.*, 1997; *Pöschl et al.*, 2000] are required to address this issue.

[15] The models vary in maturity; most have undergone some previous validation studies, but not always in their current configurations (see Appendix A). The main purpose of this paper is not model validation, rather we have made the general assumption that the models produce reasonable simulations of the key chemical species. We check this by making a comparison with ozonesonde measurements (below). Other publications in this wider ACCENT study more comprehensively validate NO<sub>2</sub> columns [*van Noije et al.*, 2006], CO (*Shindell et al.*, submitted manuscript, 2006), deposition budgets (*Dentener et al.*, submitted manuscript, 2005), and surface ozone (*Ellingsen et al.*, manuscript in preparation, 2006).

### 3. Description of the Model Experiments

[16] Five model simulations were specified (Table 1): a year 2000 base case (S1), three 2030 emissions cases (S2–S4, driven by the same meteorological data as S1), and a repeat of S2, but driven by a 2030s climate (S5). Simulations performed by individual models are reported in Table 2. Some models performed multiannual simulations; for these models, results were averaged over all years, to reduce the effects of interannual variability. Multiannual simulations are crucial for isolating the impact of climate change (S5–S2), but are less important for assessing the impacts of emissions changes. Two models (STOCHEM\_HadAM3 and UM\_CAM) performed multiannual simulations for S5 and S2, but just single year simulations for the other cases; where differences are calculated for these models, we compare like with like (i.e., two single year simulations or two multiyear simulations). Spin-ups of at least 3 months were used for all experiments.

#### 3.1. Emissions

[17] Gridded 1° × 1° anthropogenic emissions of NO<sub>x</sub>, CO, NMHCs, SO<sub>2</sub> and NH<sub>3</sub> were specified for the year 2000 and three future emissions scenarios; global totals are given in Table 3. To save time spinning up models, and to help constrain the results, we specified global methane mixing ratios across the model domain (Table 3), using results from earlier transient (1990–2030) integrations of STOCHEM\_HadAM3 [*Dentener et al.*, 2005; *Stevenson et al.*, 2005], together with Intergovernmental Panel on Climate Change (IPCC) recommendations for A2 [*IPCC*, 2001, Table II.2.2]. Year 2000 (S1) emissions are based on the EDGAR3.2 data set [*Olivier and Berdowski*, 2001]; three 2030 scenarios are S2, IIASA “current legislation” (CLE); S3, IIASA “maximum feasible reductions” (MFR) [*Dentener et al.*, 2005]; and S4, IPCC SRES A2

**Table 2.** Number of Simulated Years (Excluding Spin-up) Performed by Individual Models<sup>a</sup>

Code Letter	Model Name	S1	S2	S3	S4	S5
A	CHASER_CTM	1	1	1	1	–
B	CHASER_GCM	9	9	–	–	9
C	FRSGC/UCI	1	1	1	1	–
D	GEOS-CHEM	1	1	1	1	–
E	GISS	10(5)	10(5)	5	5	10
F	GMI/CCM3	1	1	1	1	–
G	GMI/DAO	1	1	1	1	–
H	GMI/GISS	1	1	1	1	–
I	IASB	1	1	1	1	–
J	LLNL-IMPACT	1	1	1	1	–
K	LMDz/INCA-CTM	1	1	1	1	–
L	LMDz/INCA-GCM	5	5	–	–	5
M	MATCH-MPIC/ECMWF	1	1	1	1	–
N	MATCH-MPIC/NCEP	1	1	1	1	–
O	MOZ2-GFDL	2	2	2	2	–
P	MOZART4	2	2	2	2	2
Q	MOZECH	5	5	–	–	5
R	MOZECH2	5	5	–	–	5
S	p-TOMCAT	1	1	1	1	–
T	STOCHEM-HadAM3	10(1)	10(1)	1	1	6
U	STOCHEM-HadGEM	5	5	5	5	5
V	TM4	1	1	1	1	–
W	TM5	1	1	–	–	–
X	UIO_CTM2	1	1	1	1	–
Y	ULAQ	10	10	10	10	10
Z	UM_CAM	10(1)	1	1	1	10

<sup>a</sup>Three models (GISS, STOCHEM-HadAM3, and UM\_CAM) performed multiannual simulations to compare with S5 as well as simulations for a smaller number of years to compare with S3 and S4.

[Nakicenovic *et al.*, 2000]. Ship emissions were added to all emissions scenarios, using EDGAR3.2 emissions for 1995 [Olivier and Berdowski, 2001], and assuming 1.5%/year growth to 2030 in all cases (except for S4, where ships were included at year 2000 levels in error; a separate experiment evaluating 2030 ship emissions for S4 is ongoing (Eyring *et al.*, Multi-model simulations of the impact of international shipping on atmospheric chemistry and climate in 2000 and 2030, manuscript in preparation, 2006)). Aircraft NO<sub>x</sub> emissions totals (Table 3) and distributions from NASA or ANCAT [Henderson *et al.*, 1999] were recommended. Aircraft NO<sub>x</sub> emissions totals for 2030 (Table 3) were based on the IS92a base scenario [Henderson *et al.*, 1999, Table 9–19]. All the 2030 emissions were “harmonized” to the 2000 emissions, by calculating changes in emissions from 2000 to 2030, then adding them to the base case emissions, to avoid minor inconsistencies.

[18] Satellite-derived monthly varying biomass burning gridded (1° × 1°) distributions [van der Werf *et al.*, 2003]

**Table 3.** Specified Global Annual Anthropogenic Emission Totals for Each Scenario<sup>a</sup>

	S1	S2	S3	S4	S5	TAR
NO <sub>x</sub> , Tg N	27.8	32.8	13.1	54.6	32.8	33
CO, Tg	470	397	222	761	397	650
NMVOC, Tg	116	114	73	176	114	161
SO <sub>2</sub> , Tg S	54	57	17	100	57	76
NH <sub>3</sub> , Tg N	49	65	65	69	65	36
CH <sub>4</sub> , ppbv	1760	2088	1760	2163	2012	1745

<sup>a</sup>Biomass burning emissions did not vary between scenarios. Specified methane mixing ratios are also given; these were fixed throughout the model domain. Ship emissions are included (for S4, ship emissions for 2000 were used in error). Additional aircraft emissions of 0.8 Tg N (S1) and 1.7 Tg N (all 2030 cases) were recommended. Values from IPCC TAR for year 2000 are also shown for comparison.

and totals (Table 4) were specified. These values are averages for the time period 1997–2002, and cover all types of fires (wildfires, savannah burning, and some agricultural waste burning). Ecosystem-specific emission factors from *Andreae and Merlet* [2001] are employed. Domestic and industrial uses of biomass as fuel are included in the anthropogenic emissions. Given the lack of information regarding future levels of biomass burning [e.g., Nakicenovic *et al.*, 2000], these values and distributions were used for 2000 and all 2030 scenarios. The SRES A2 scenario anthropogenic emissions include some biomass burning emissions; these were removed [see, e.g., Stevenson *et al.*, 2000] to avoid double counting. Full details of the anthropogenic and biomass burning emissions can be found at (<http://ccupeople.jrc.it/dentener/index.htm>). Other emissions were specified by individual modelers.

[19] Lightning NO<sub>x</sub> production in many of the models is directly linked to the distribution of convection; this is probably the most important and most uncertain natural source of NO<sub>x</sub> [e.g., Price *et al.*, 1997; Pickering *et al.*, 1998]. Modelers used lightning NO<sub>x</sub> emissions in the range 3.7–7.0 Tg(N) yr<sup>−1</sup>; in the central part of the range suggested by Price *et al.* [1997] (1–20 Tg(N) yr<sup>−1</sup>). Soil NO<sub>x</sub> emissions also represent an important natural source, augmented by fertilizer application [e.g., Yienger and Levy, 1995]. Modelers used values in the range 5.5–8.0 Tg(N) yr<sup>−1</sup>.

[20] Biogenic emissions of isoprene represent the largest NMHC source, mainly emitted from tropical forests. One estimate of total annual emission is 503 Tg(C) [Guenther *et al.*, 1995]. As for the treatment of lightning, some models include interactive isoprene emissions. Modelers used values in the range 220–631 TgC/yr. This relatively wide range, combined with differences in isoprene oxidation mechanisms, will contribute to differences in ozone, particularly in the tropics. Some models also emit significant quantities of other biogenic compounds (e.g., MOZ2-GFDL: Terpene 260 TgC/yr; CH<sub>3</sub>OH 287 Tg/yr).

[21] Models include NMHC schemes of varying complexity. The speciation of NMHCs in anthropogenic and biomass burning emissions given by Prather *et al.* [2001, Table 4.7] was recommended, with species not included in the model either ignored or lumped into related species (e.g., higher alkanes were included as butane). Differences

**Table 4.** Specified Global Biomass Burning and Recommended Natural Emissions Sources<sup>a</sup>

	Sources	This Study	IPCC TAR
NO <sub>x</sub> , Tg N	biomass burning	10.2	7.1
NO <sub>x</sub> , Tg N	soils	7	5.6
NO <sub>x</sub> , Tg N	lightning	5	5.0
CO, Tg	biomass burning	507	700
CO, Tg	oceans/vegetation	100	200
NMHC, Tg	biomass burning	31	42
NMHC, Tg	vegetation isoprene	580	249
NMHC, Tg	vegetation terpenes	295	144
SO <sub>2</sub> , Tg S	biomass burning	1.4	2.2
SO <sub>2</sub> , Tg S	volcanoes	14.6	9.3
DMS, Tg S	oceans/terrestrial biosphere	20	24
NH <sub>3</sub> , Tg N	biomass burning	4.9	5.7
NH <sub>3</sub> , Tg N	oceans	8.3	8.2
NH <sub>3</sub> , Tg N	soils	2.4	2.4

<sup>a</sup>Fixed for all scenarios, except in some models for S5, where some natural emissions are linked to climate. Values from IPCC TAR are shown for comparison.

in model NMHC emission magnitudes, speciation, and chemical schemes probably lead to important variations in modeled ozone, because of the variable formation of PAN and other important  $\text{NO}_y$  reservoir species. Additional natural sources of trace gases were also recommended (Table 4).

[22] Most models added emissions to the lowest model layer; however several sources (e.g., large industrial sources, major wildfires, and aircraft) inject emissions at higher altitudes. All the models included 3-D emissions of aircraft  $\text{NO}_x$ . Height profiles for different emission sectors were recommended for those models with the capability to add emissions over several layers. Some models (GMI, IASB, TM4, and TM5) added industrial emissions over the height range 100–300 m; these models and four others (GFDL-MOZ2, LLNL-IMPACT, MOZECH and MOZECH2) implemented biomass burning emissions specified in six layers (boundaries at 0, 0.1, 0.5, 1, 2, 3, and 6 km) [Dentener *et al.*, 2006b].

### 3.2. Climate Change Scenario (S5)

[23] To gauge the impact of climate change on tropospheric composition by 2030, we repeated the midrange emissions case (S2), but changed the underlying climate to one appropriate for 2030. To generate more statistically significant results, multiannual (5–10 years) simulations were performed and averaged, in order to reduce confounding signals originating from interannual climate variability. The methane boundary condition for S5 was reduced relative to S2 (Table 3), based on earlier results from a fully transient simulation over the time period 1990–2030 using the STOCHEM-HadAM3 model [Stevenson *et al.*, 2005]. Methane concentrations are lower when climate change is taken into account because higher temperatures are accompanied by higher humidities and hence higher OH levels. Higher temperatures and OH levels both promote methane oxidation.

[24] All of the GCMs were configured as atmosphere-only models, with prescribed sea surface temperatures (SSTs) providing the lower boundary condition over the oceans. SSTs and sea-ice distributions from an existing simulation of HadCM3 (Hadley Centre Coupled Model, version 3 [Johns *et al.*, 2003]), forced by the IS92a scenario [Leggett *et al.*, 1992; Cox *et al.*, 2000] were used by most models for the 2030 climate. Some models used their own climate simulations under different scenarios (e.g., MOZECH, MOZECH2: B1; STOCHEM\_HadGEM: A2). LMDz/INCA-GCM used SSTs from Rayner *et al.* [1996] for year 2000, together with changes to 2030 given by [Dufresne *et al.*, 2002]. The use of a range of climate scenarios for 2030 introduces a range in the magnitude of the applied climate change; another source of difference between GCMs arises from differing climate sensitivities [e.g., Cubasch *et al.*, 2001]. Global annual mean surface warming between 2000 and 2030 varied from 0.31 K (MOZECH2) to 0.95 K (CHASER\_GCM). The range of climate change and chemical responses are discussed in more detail below.

## 4. Results and Discussion

[25] Each model reported 3-D monthly mean ozone mixing ratios, ozone budget terms and methane oxidation fluxes on their native grids. Each model also reported the mass of each grid box, allowing global burdens and fluxes

to be easily computed. The focus of this study was the troposphere; we apply a mask for grid boxes where monthly mean  $\text{O}_3 > 150$  ppbv [e.g., Prather *et al.*, 2001], effectively removing regions above the tropopause. We apply a consistent mask for each model across all scenarios, normally using the ozone field from S1 but in a few cases using the maximum grid box value across all scenarios (this method was used for GMI and GEOS-CHEM, as they masked results “online,” and this was needed to create a consistent mask across all scenarios for these models). Previous studies [e.g., Stevenson *et al.*, 2004] have shown that using various tropopause definitions (e.g., a fixed pressure level, the dynamical tropopause, or the chemical tropopause) can result in significant differences for some terms, such as the tropospheric ozone lifetime and burden. These differences hinder comparisons between models. By using a consistent definition for all models we hope to reduce this source of model differences.

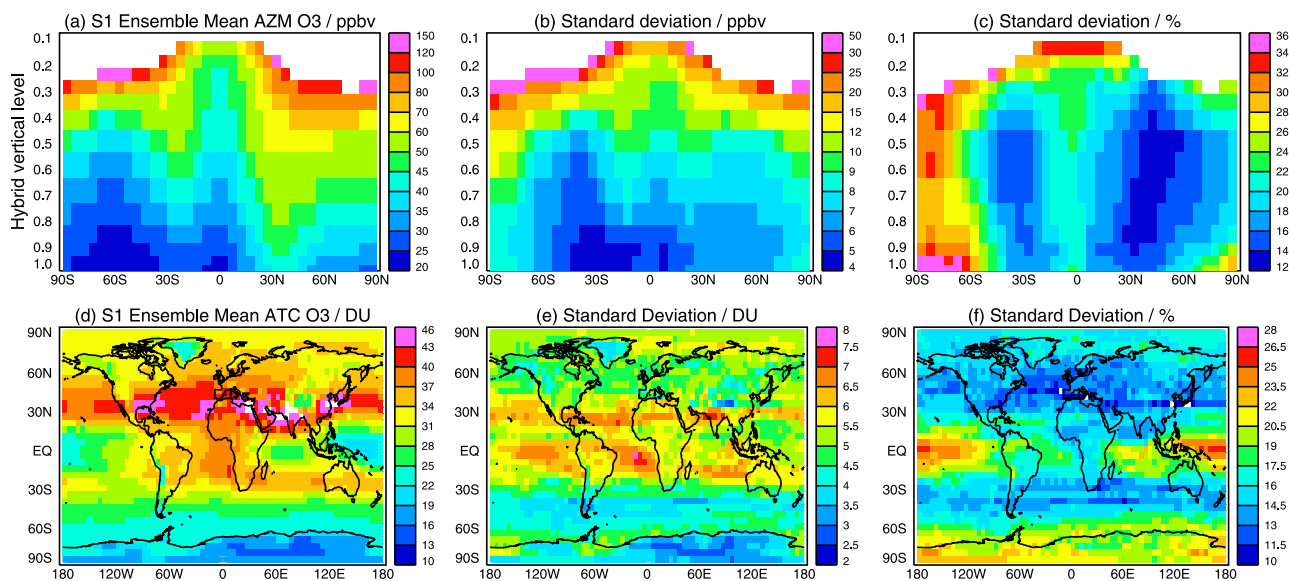
[26] Models used a wide variety of vertical coordinate systems and resolutions. Ozone results were converted to a common vertical grid: the 19 hybrid (sigma pressure) levels of the Met. Office HadAM3 model (the native vertical grid of the UM\_CAM model); up to 14 of these levels span the troposphere. Results were also interpolated to a common horizontal resolution of  $5^\circ \times 5^\circ$ . With results on a common grid, ensemble means and standard deviations for each grid box were calculated. Ozone fields on this grid were also used to calculate radiative forcings. For global tropospheric burdens and budgets, model results were masked (as described above) and then summed on their native grids, to avoid the introduction of any minor errors associated with interpolation.

### 4.1. Ozone Distributions

#### 4.1.1. Base Year 2000 (S1)

[27] Figure 1 shows ensemble means and standard deviations (absolute and percentage) of annually averaged zonal mean and tropospheric column ozone. The ensemble comprises all 26 models (Table 2). Use of an ensemble should improve the robustness of model results, as individual model errors are likely to cancel, whereas the real signal should reinforce [e.g., Cubasch *et al.*, 2001; Pagowski *et al.*, 2005]. The distribution of standard deviations (SDs) indicates where the models are most consistent, and where they are most uncertain. High SDs close to the tropopause (Figure 1b) are unsurprising, as the models will, almost inevitably, place this region of rapid change in ozone mixing ratios at slightly different altitudes. High SDs throughout the polar troposphere (particularly over the Antarctic; Figures 1c and 1f) are probably also related to differences in the tropopause level at these latitudes, and the resultant stratospheric injection of ozone to the troposphere. Perhaps the most interesting aspect of Figures 1c and 1f is the relatively high SDs ( $\sim 30\%$ ) in the tropics compared to relatively low SDs at midlatitudes ( $\sim 20\%$ ). This larger model uncertainty in the tropics perhaps reflects important intermodel differences in convection parametrizations, biogenic isoprene emissions and chemistry, lightning  $\text{NO}_x$  production, and the treatment of biomass burning emissions [e.g., Peters *et al.*, 2002]. Midlatitudes are less affected by these factors, and are also closer to the majority of emissions sources; however they might be expected to be more





**Figure 1.** Modeled ozone from case S1 (year 2000): (a–c) zonal annual means (ppbv) and (d–f) tropospheric columns (DU). Figures 1a and 1d are ensemble means (all 26 models); Figures 1b and 1e are absolute standard deviations; Figures 1c and 1f are percentage standard deviations (also known as coefficient of variation). To calculate the ensemble values, individual model results were interpolated to a common grid ( $5^\circ \times 5^\circ \times 19$  levels) and masked at the chemical tropopause ( $O_3 = 150$  ppbv).

susceptible to uncertainties in stratosphere-troposphere exchange, particularly in the upper troposphere.

[28] The column  $O_3$  plots also show the largest SDs in the tropics, particularly over the oceans. The “step change” in SDs at around  $30^\circ\text{N}/30^\circ\text{S}$  (Figure 1e) reflects the latitude of the subtropical jet, where the tropopause shows a discontinuity and jumps to higher levels in the tropics [Highwood and Hoskins, 1998]. Because models place this boundary at slightly different latitudes, this heightens uncertainties in this region. The largest absolute uncertainties are associated with the ozone peak over the tropical South Atlantic (Figure 1e); this peak is seen in observations [e.g., Thompson *et al.*, 2003a, 2003b], and is thought to be associated with the outflow of lightning and biomass burning emissions from Africa. The largest relative uncertainties are for the low-ozone columns over the tropical Pacific (Figure 1f); the large range may relate to model variations in the efficiency of ozone destruction chemistry in comparatively clean environments.

#### 4.1.2. Comparison With Ozone Sonde Data

[29] Figure 2 compares the model ensemble mean  $O_3$  for year 2000 with ozonesonde observations, taken from Logan [1999] and Thompson *et al.* [2003a, 2003b]. The Logan data are representative of the time period 1980–1993, while the Thompson *et al.* data are for the years 1997–2002. Although the Logan data do not coincide in time with our simulated year (2000), preliminary analysis of more recent global ozonesonde data reveal only minor trends, suggesting that the earlier data are a useful source for comparison (J. Logan, personal communication, 2005). The Logan [1999] sonde data are located mainly in northern midlatitudes over continental North America, Europe, and Japan, but have been significantly supplemented by the Southern Hemisphere Additional Ozonesondes (SHADOZ) data in the southern tropics [Thompson *et al.*, 2003a, 2003b]. The northern

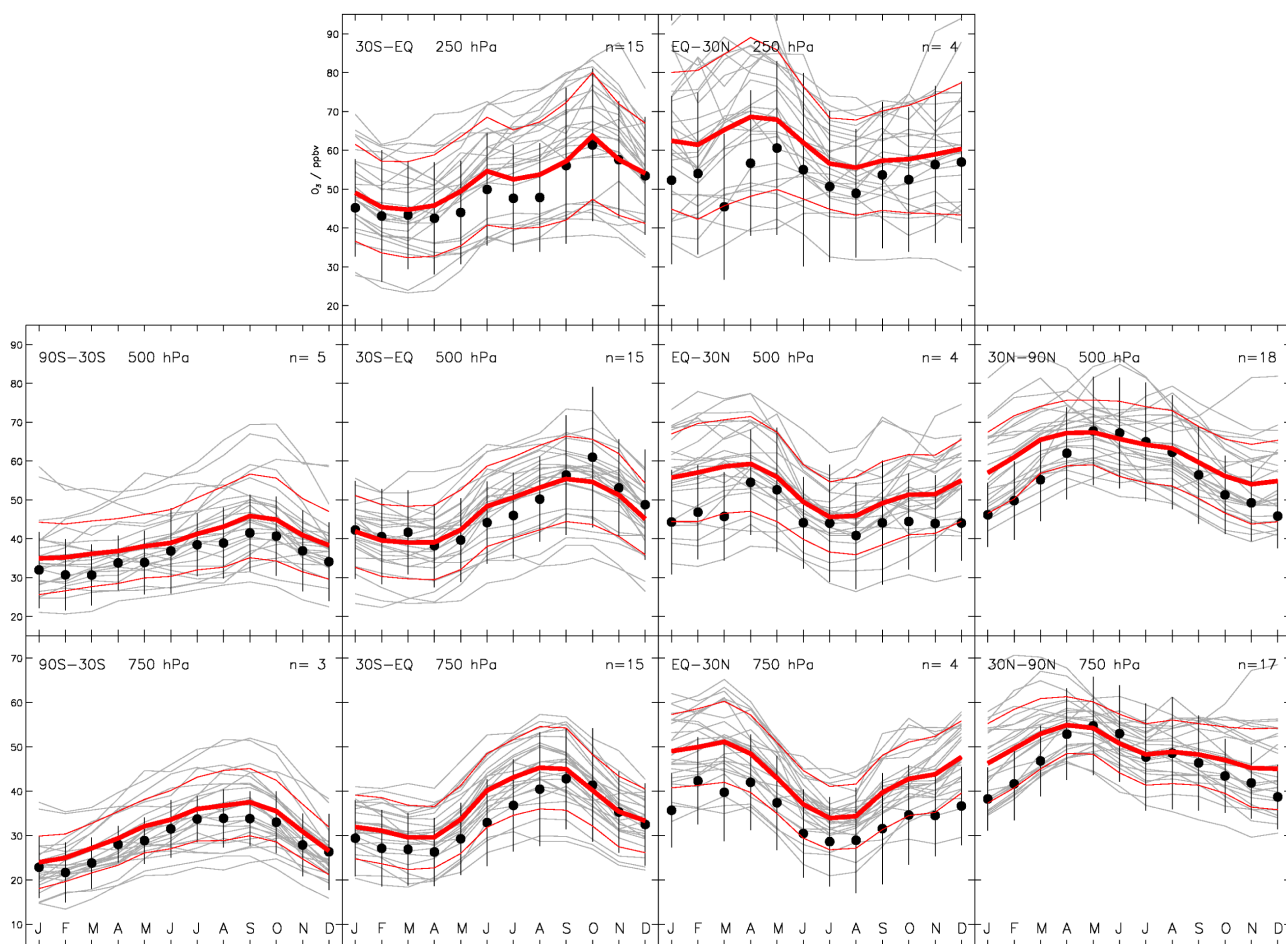
tropics and southern midlatitudes are underrepresented (Figure 2 indicates the number of sites used for each plot).

[30] The comparison indicates that the mean model closely resembles the observations, with the mean values nearly always within a standard deviation of each other. The mean model tends to underestimate the amplitude of the seasonal cycle at  $30^\circ$ – $90^\circ\text{N}$ , overestimating winter ozone by around 10 ppbv. This may reflect the lack of a seasonal cycle in anthropogenic emissions (although it is not obvious that this would improve results), or indicate deficiencies in descriptions of NH stratospheric influx of  $O_3$ . The mean model also tends to slightly overpredict ozone throughout the northern tropics, although this region is only represented by four sites (Naha, Japan; Hilo, Hawaii; Poona, India; and Paramaribo, Suriname). Comparisons in the Southern Hemisphere show good agreement, suggesting that the models’ representation of biomass burning and natural processes (such as lightning and soil emissions) are quite realistic.

[31] Figure 2 also shows results for all individual models (gray lines). These are shown for each model in the auxiliary material.<sup>1</sup> Taking each monthly mean point at each vertical level shown in Figure 2, for each model we calculated a root-mean-square error (RMSE) of the simulated ozone compared to the observed mean. The range in RMSE across all the models is 5.1 to 18.0 ppbv; the mean model has an RMSE of 5.9 ppbv. We later use these RMSE values as part of a criterion for checking the influence of the outlying models on the mean results; models with RMSE values in excess of 12.5 ppbv are considered outliers.

[32] Further validation of ozone precursors is required to increase our confidence in the models’ abilities;  $\text{NO}_2$  columns and CO are considered in related papers [van Noije *et al.*, 2006; Shindell *et al.*, submitted manuscript,

<sup>1</sup>Auxiliary material is available at <ftp://ftp.agu.org/apend/jd/2005jd006338>.



**Figure 2.** Comparison of the annual cycle of ozone observations (black dots) and model ensemble mean (thick red line), sampled for different latitude bands ( $90^{\circ}\text{S}$ – $30^{\circ}\text{S}$ ,  $30^{\circ}\text{S}$  to equator; equator to  $30^{\circ}\text{N}$ , and  $30^{\circ}\text{N}$ – $90^{\circ}\text{N}$ ) and different pressure levels (750, 500, and 250 hPa). The gray lines are the results for each model (also see individual plots in the auxiliary material). Each panel is the mean of several sites (the number of sites is given in the top right corner of each plot); the model and observations are sampled in exactly the same way. The bars for each observation are the average of the interannual standard deviations at each station. The thin red lines are the standard deviation of the 26 model ensemble. These standard deviations therefore represent different measures of uncertainty, but indicate approximate errors for each point. Data are from Logan [1999] and Thompson *et al.* [2003a, 2003b].

2006]. Validation of hydrocarbons is also highly desirable, but is beyond the scope of this paper. Surface ozone from these simulations is validated in detail by Ellingsen *et al.* (manuscript in preparation, 2006).

#### 4.1.3. Changes 2030–2000 Under Different Emissions Scenarios (S2, S3, and S4)

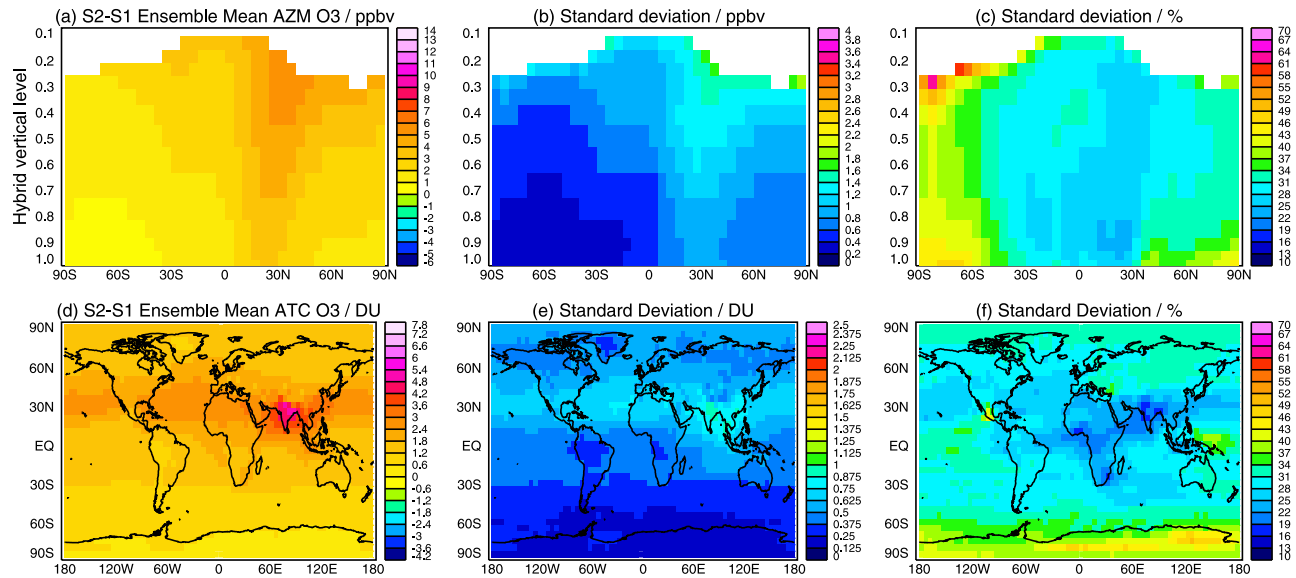
[33] Figures 3, 4, and 5 present results for the differences in ozone between the three 2030 scenarios (S2, S3 and S4) and the year 2000, following the same format as Figure 1. Under the CLE scenario (S2), ensemble mean ozone shows a zonal annual mean increase of up to 6 ppbv in the northern subtropical upper troposphere (Figure 3a), with an intermodel standard deviation of typically 20–30% (Figure 3c). Changes in column ozone show that the peak increase occurs over India (Figure 3d), reflecting the lack of current legislation limiting ozone precursor emissions in this region.

[34] In the MFR scenario (S3) ozone decreases throughout the troposphere (Figure 4), by up to 6 ppbv in the annual

zonal mean, with peak decreases occurring in the lower troposphere at  $30^{\circ}$ – $40^{\circ}\text{N}$ . These changes have an associated uncertainty of order 20–30% (similar to those in S2), but there are also significant changes in the upper troposphere with a larger uncertainty of 40–50% (Figure 4c). The larger uncertainties compared to the CLE scenario reflect the more widespread significant changes in emissions (over the entire industrialized world) under this scenario; this experiment therefore samples a wider cross section of model processes. Changes in column ozone (Figure 4d) highlight the industrial regions, with some weighting toward the equator; these locations are those most susceptible to technological reductions in emissions, and hence ozone.

[35] Results for the SRES A2 scenario (S4, Figure 5) remind us of what would happen if we choose to ignore current legislation and allow large growth in ozone precursor emissions; zonal annual mean ozone increases everywhere, typically by 6–10 ppbv in the Northern Hemisphere,



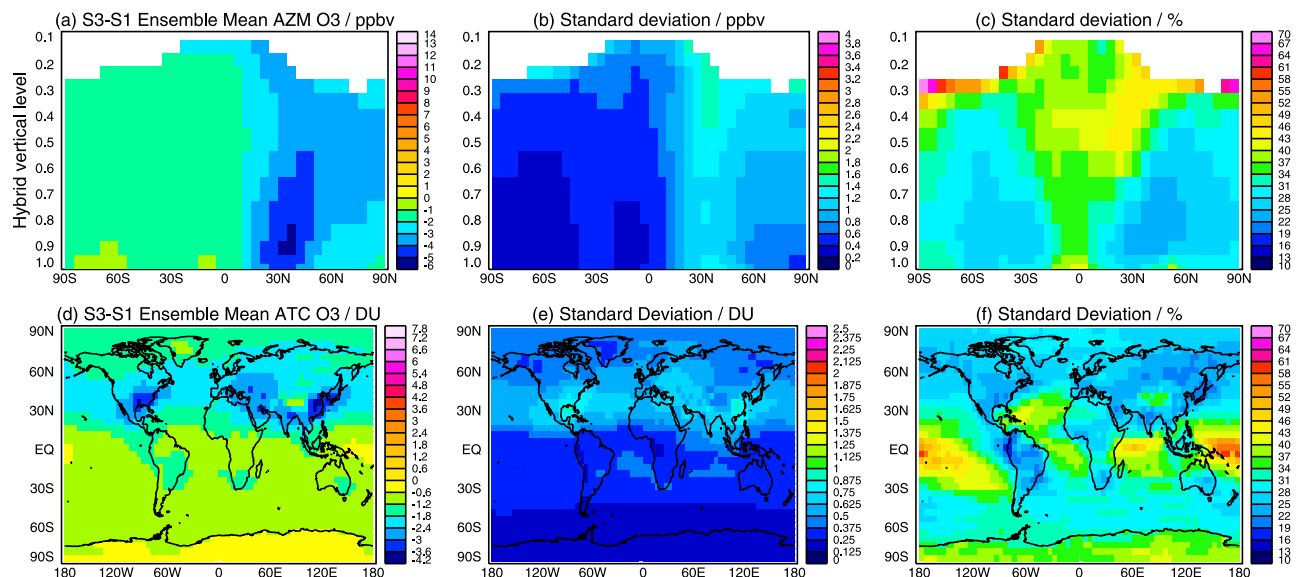


**Figure 3.** Following the same layout as for Figure 1, the ensemble mean and standard deviation modeled ozone change between case S2 (CLE 2030) and S1 (year 2000). The ensemble comprises all 26 models. Figures 3–5 share the same scales for all panels.

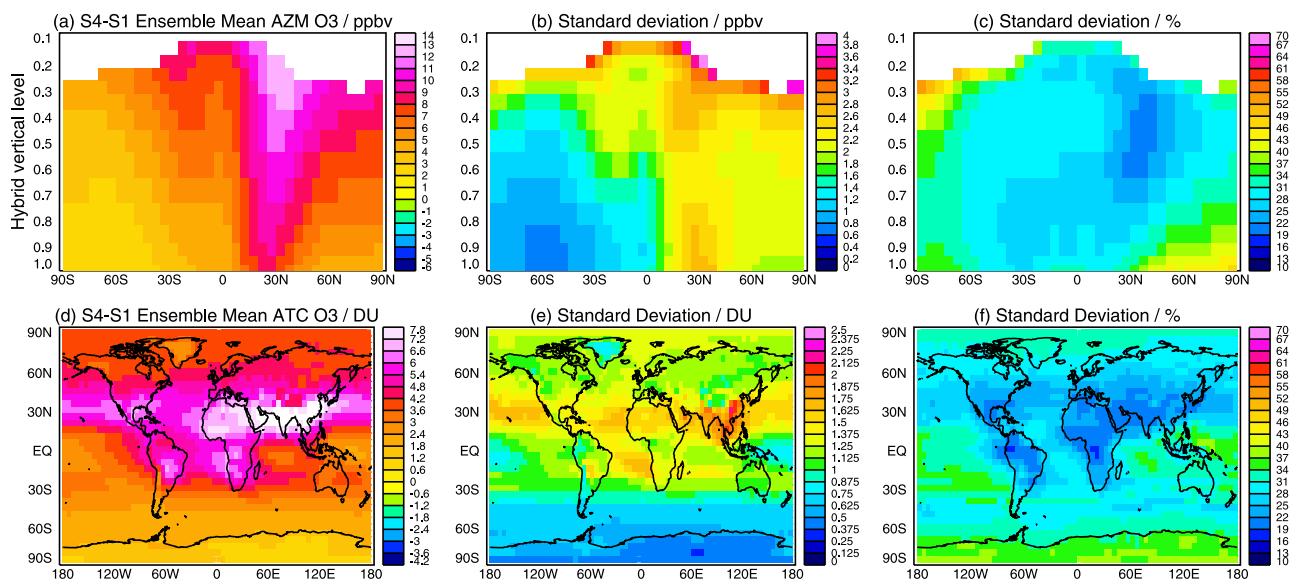
and by up to 13 ppbv in the northern subtropical upper troposphere. The changes have an intermodel uncertainty of around 20–30%, over much of the troposphere (also similar to S2). The column ozone changes show the widespread growth of ozone, particularly in the rapidly developing world (China, southeast Asia, Middle East, plus parts of Africa and Latin America).

[36] The changes in tropospheric ozone burden associated with these three scenarios show a broadly linear relation to changes in  $\text{NO}_x$  emissions (Figure 6). The emission

changes for scenarios S2–S4 relative to S1 represent changes in total emitted  $\text{NO}_x$  of +12%, –27% and +55% respectively, and they yield changes in  $\text{O}_3$  burden of +6%, –5%, and +15%. A linear relationship should not be expected, partly because of the nonlinearities in  $\text{O}_3$  chemistry, but also because of the influence of changes in other trace gas emissions, such as  $\text{CO}$ ,  $\text{CH}_4$  and NMHCs [e.g., Wang and Jacob, 1998]. In addition, the relatively meager reduction in ozone under the MFR scenario partly reflects that aircraft emissions show the same growth to 2030 in all



**Figure 4.** Following the same layout as for Figure 3, the ensemble mean and standard deviation modeled ozone change between case S3 (MFR 2030) and S1 (year 2000). The ensemble comprises the 21 models indicated in Table 2. Figures 3–5 share the same scales for all panels.



**Figure 5.** Following the same layout as for Figure 3, the ensemble mean and standard deviation modeled ozone change between case S4 (A2 2030) and S1 (year 2000). The ensemble comprises the 21 models indicated in Table 2. Figures 3–5 share the same scales for all panels.

scenarios, and although the total aircraft emission is small compared to total  $\text{NO}_x$ , it has a disproportionately large impact, especially as surface emissions are dramatically reduced. This highlights the future problems related to unregulated growth of the aircraft sector, particularly under optimistic scenarios for surface emissions. Ship emissions present similar problems. Nevertheless, Figure 6 provides our best estimate of the overall relationship between changes in global  $\text{NO}_x$  emissions and tropospheric ozone.

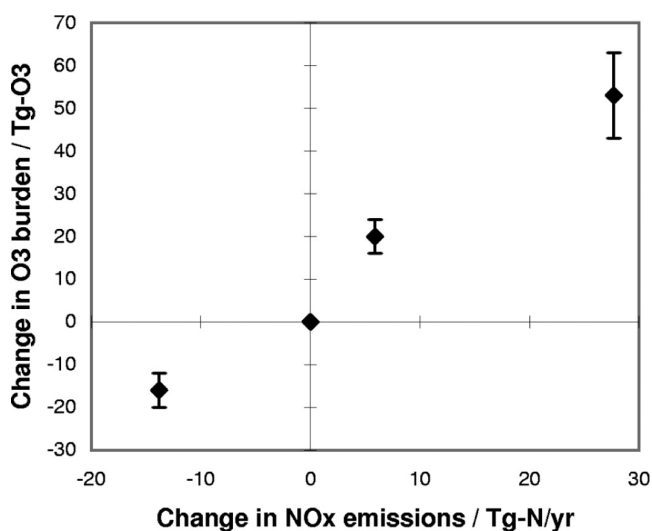
#### 4.1.4. Influence of Climate Change

[37] Figure 7 shows changes in ozone between S5 and S2; Figure 7 follows a slightly different format to Figures 3–5 because the intermodel differences are larger and range between increases and decreases in ozone. For this reason, we plot the ensemble mean (Figures 7b and 7e) together with the mean plus/minus 1 standard deviation, to illustrate the range in results. The mean tropospheric  $\text{O}_3$  burden shows a slight decrease related to climate change, with the decrease mainly in the lower troposphere, particularly over the tropical oceans. This negative climate feedback appears to be related to higher humidities and hence increased ozone destruction via reaction (R3) (see section 4.2). At the same time, upper tropospheric ozone rises, especially in the Northern Hemisphere, which is related to an increased influx from the stratosphere. The opposite change occurs in the Southern Hemisphere, owing to a reduced stratospheric influx. These two climate feedbacks (water vapor and stratospheric input) appear to be the dominant mechanisms operating, and different models show variable sensitivities. The two STOCHEM models and GISS are dominated by the water vapor feedback; the other models also tend to show some reductions in tropical lower tropospheric ozone, but are also variably influenced by changes in stratospheric input. We should note that a small reduction in ozone originates from the prescribed change in

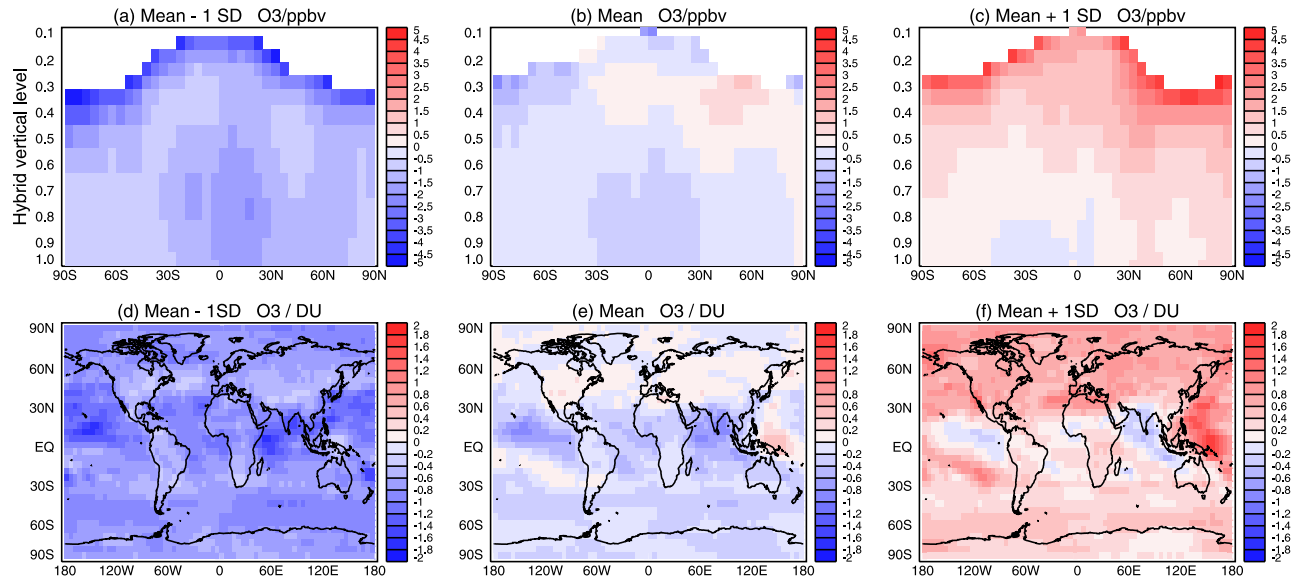
methane mixing ratios (Table 3). This prescribed change comes from earlier, transient runs of the STOCHEM\_HadAM3 model, over the time period 1990–2030, using very similar emissions scenarios. The response of methane lifetime in this model is, reassuringly, broadly consistent with others, suggesting that fully transient experiments with all models would produce similar results.

#### 4.2. Ozone Budgets

[38] Table 5 gives year 2000 (S1) tropospheric ozone budgets for individual models, together with an ensemble



**Figure 6.** Ensemble mean change in annual mean tropospheric ozone burden versus change in global  $\text{NO}_x$  emissions for the three scenarios, S2, S3, and S4, relative to S1. The bars represent  $\pm 1$  standard deviation within the ensemble members.



**Figure 7.** Ozone changes due to climate change: the difference between case S5 (CLE 2030, with a year 2030 climate) and S2 (CLE 2030, with a year 2000 climate). The standard deviations are larger for this case, and the modeled range of ozone change varies in sign. (a and d) Ensemble mean minus 1 standard deviation; (b and e) ensemble mean change; (c and f) ensemble mean plus 1 standard deviation. The ensemble comprises the 10 models indicated in Table 2.

**Table 5.** Tropospheric O<sub>3</sub> Budget, Burden, Lifetime, and Methane Lifetime for the S1 Simulation for Individual Models<sup>a</sup>

	P	L	D	S <sub>inf</sub>	B <sub>O<sub>3</sub></sub>	τ <sub>O<sub>3</sub></sub>	τ <sub>CH<sub>4</sub></sub>
<b>A. CHASER_CTM</b>	5042	4594	948	501	331	21.8	8.42
<b>B. CHASER_GCM</b>	5032	4620	948	536	333	21.8	8.37
<b>C. FRSGC/UCI</b>	5135	4733	907	505	331	21.4	7.61
D. GEOS-CHEM	4490	3770	1016	296	294	22.4	10.17
<b>E. GISS</b>	—	—	—	—	341	—	8.48
F. GMI/CCM3	5331	5059	862	590	388	23.9	7.50
G. GMI/DAO	5124	4940	763	579	386	24.7	7.64
<b>H. GMI/GISS</b>	4722	4396	856	530	372	25.9	8.54
I. IASB	—	—	—	—	377	—	8.12
J. LLNL-IMPACT	5432	5160	1014	742	406	24.0	7.18
<b>K. LMDz/INCA-CTM</b>	4912	4182	1232	502	330	22.3	8.57
<b>L. LMDz/INCA-GCM</b>	4931	4027	1227	324	316	22.0	8.78
M. MATCH-MPIC/ECMWF	—	—	—	—	377	—	—
N. MATCH-MPIC/NCEP <sup>a</sup>	4342 <sup>a</sup>	4324 <sup>a</sup>	948 <sup>a</sup>	930 <sup>a</sup>	399	27.9 <sup>a</sup>	9.48
<b>O. MOZ2-GFDL</b>	5263	5087	963	787	349	21.0	8.42
<b>P. MOZART4</b>	4964	4670	906	612	375	24.5	9.07
Q. MOZECH	6920	6617	963	660	407	19.6	6.31
R. MOZECH2	6130	5876	925	671	387	20.7	7.16
S. p-TOMCAT	—	—	—	—	248	—	12.46
<b>T. STOCHEM-HadAM3</b>	5331	4821	945	435	274	17.3	8.44
U. STOCHEM-HadGEM	5114	3757	1507	151	293	20.3	10.36
<b>V. TM4</b>	4806	4594	720	508	344	23.6	8.80
<b>W. TM5</b>	4580	4623	827	871	339	22.7	7.93
X. UIO_CTM2	—	—	—	—	—	—	10.33
Y. ULAQ	5009	4469	1356	623	328	21.3	8.06
Z. UM_CAM	3922	3363	1172	614	303	24.4	10.57
Mean ± standard deviation (all models)	5110 ± 606	4668 ± 727	1003 ± 200	552 ± 168	344 ± 39	22.3 ± 2.0	8.67 ± 1.32
Mean ± standard deviation (selected models)	<b>4974 ± 223</b>	<b>4577 ± 291</b>	<b>953 ± 154</b>	<b>556 ± 154</b>	<b>336 ± 27</b>	<b>22.2 ± 2.2</b>	<b>8.45 ± 0.38</b>
IPCC TAR	3420	3470	770	770	300	24	8.4

<sup>a</sup>P is chemical production, L is chemical loss, D is surface deposition, and S<sub>inf</sub> is stratospheric input – inferred as the residual of the other terms (all in Tg(O<sub>3</sub>) yr<sup>-1</sup>); B<sub>O<sub>3</sub></sub> is burden (Tg(O<sub>3</sub>)), τ<sub>O<sub>3</sub></sub> is lifetime (days), and τ<sub>CH<sub>4</sub></sub> is methane lifetime (years, for the whole atmosphere, assuming a soil sink of 30 Tg yr<sup>-1</sup> and a stratospheric sink of 40 Tg yr<sup>-1</sup>). Models with no O<sub>3</sub> budget terms are not used to calculate the mean values for any of the O<sub>3</sub> terms; budgets for model N were also excluded. Budget terms from model N were calculated using a WMO tropopause definition. Values in italics are > 1 SD above or below the mean value. Mean values are also calculated using a subset of the models, shown in bold (see text for selection criteria). Mean values from IPCC TAR are also given for comparison.



**Table 6.** Interscenario Ensemble Mean Changes in Tropospheric O<sub>3</sub> Budget Terms, Burden, and Lifetime, and Whole Atmosphere Methane Lifetimes<sup>a</sup>

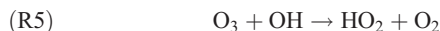
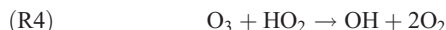
	$\Delta P$ , Tg(O <sub>3</sub> )yr <sup>-1</sup>	$\Delta L$ , Tg(O <sub>3</sub> )yr <sup>-1</sup>	$\Delta D$ , Tg(O <sub>3</sub> )yr <sup>-1</sup>	$\Delta S_{\text{inf}}$ , Tg(O <sub>3</sub> )yr <sup>-1</sup>	$\Delta B_{\text{O}_3}$ , Tg(O <sub>3</sub> )	$\Delta \tau_{\text{O}_3}$ , days	$\Delta \tau_{\text{CH}_4}$ , years
S2-S1	397 ± 66(7.8%)	325 ± 59(7.0%)	59 ± 16(5.9%)	-11 ± 20(-2.0%)	20 ± 4(5.8%)	-0.19 ± 0.09(-0.9%)	0.23 ± 0.20(2.7%)
S3-S1	-454 ± 70(-8.9%)	-342 ± 47(-7.3%)	-94 ± 28(-9.4%)	18 ± 32(3.3%)	-16 ± 4(-4.7%)	0.76 ± 0.15(3.4%)	0.12 ± 0.18(1.4%)
S4-S1	1205 ± 147(24%)	963 ± 121(21%)	205 ± 54(20%)	-36 ± 75(-6.5%)	53 ± 10(15%)	-1.1 ± 0.2(-4.9%)	0.00 ± 0.31(0.0%)
S5-S2	32 ± 62(0.6%)	87 ± 73(1.7%)	-14 ± 11(-1.3%)	41 ± 31(7.6%)	-1.4 ± 4.2(-0.4%)	-0.34 ± 0.38(-1.5%)	-0.35 ± 0.16(-3.9%)

<sup>a</sup>Mean is shown ±1 standard deviation, and percentage changes, relative to S1 for S2-S1, S3-S1, and S4-S1 and relative to S2 for S5-S2, are shown in parentheses.

mean and standard deviation. Chemical production (P) is defined as the sum of all the major reactions that convert NO to NO<sub>2</sub>:



Chemical loss (L) is dominated by the reactions:



A further small contribution comes from the reactions of O<sub>3</sub> with alkenes and NO<sub>x</sub>. Finally, minor net O<sub>3</sub> destruction occurs via a large array of other reactions and processes; we recommended following a scheme whereby net losses of O(<sup>1</sup>D), O(<sup>3</sup>P), NO<sub>2</sub> or PAN, in addition to O<sub>3</sub>, were considered as net ozone losses. In this scheme HNO<sub>3</sub>, NO<sub>3</sub> and N<sub>2</sub>O<sub>5</sub> are ignored. Some modelers used existing schemes that operated under similar principals; the important point is that fast cycles between ozone-related species are ignored.

[39] Modelers also reported surface deposition fluxes (D). From the three terms, P, L and D, and assuming no tropospheric ozone burden trend (this was confirmed to be negligible in several models), the budget was closed by an inferred stratospheric input term (S<sub>inf</sub>):

$$S_{\text{inf}} = L + D - P \quad (1)$$

Table 5 also reports tropospheric ozone burdens (B<sub>O<sub>3</sub></sub>), and lifetimes (τ<sub>O<sub>3</sub></sub>), defined as

$$\tau_{\text{O}_3} = B_{\text{O}_3} / F_{\text{O}_3} \quad (2)$$

where F<sub>O<sub>3</sub></sub> is the “ozone turnover flux,” given by

$$F_{\text{O}_3} = L + D (= P + S_{\text{inf}}) \quad (3)$$

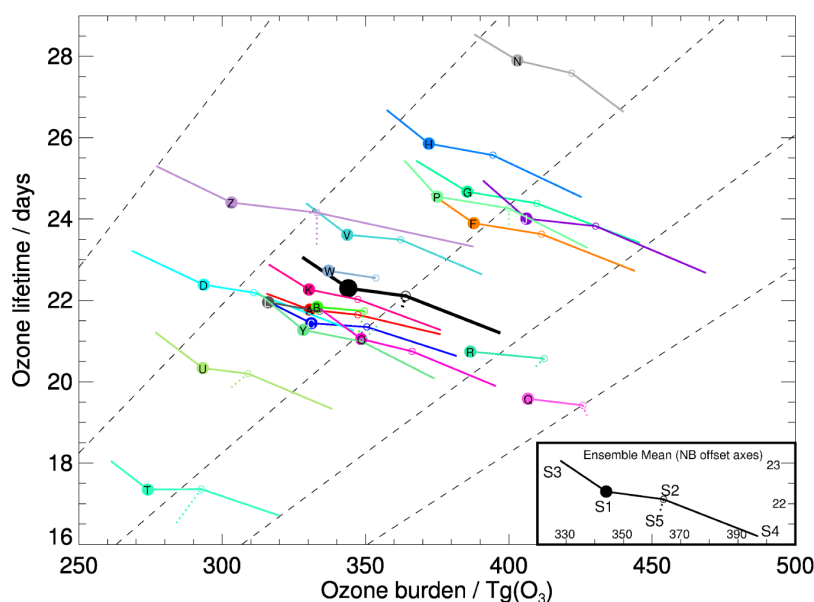
Table 5 shows mean P, L, D, and S<sub>inf</sub> terms of 5110, 4670, 1000 and 550 Tg(O<sub>3</sub>) yr<sup>-1</sup> respectively, with intermodel standard deviations of 12, 16, 20 and 30%. These can be compared to IPCC TAR values [Prather *et al.*, 2001,

Table 4.12] (also given in Table 5). It should be noted that the IPCC TAR O<sub>3</sub> budget values represent several independent model studies published between 1996 and 2000, and that these individual studies used a range of emissions, different from those recommended by TAR (reported here in Tables 3 and 4), with total NO<sub>x</sub> emissions typically 20% lower than those recommended by TAR. The year 2000 total NO<sub>x</sub> emissions recommended by TAR is similar to that used in this study. TAR also recommended isoprene emissions of 220 TgC yr<sup>-1</sup>, less than half that suggested here (Table 4). The models in TAR Table 4.12 used a wide range of isoprene emissions (some had none), but were generally lower than the emissions used in this study. Table 5 also shows mean B<sub>O<sub>3</sub></sub> of 340 Tg(O<sub>3</sub>) (±11%) and τ<sub>O<sub>3</sub></sub> of 22 days (±9%); these compare to TAR values of 300 Tg(O<sub>3</sub>) (±10%) and 24 days (±8%).

[40] The higher levels of P and L reported here (relative to TAR) are thought to be due to several reasons: (1) higher NO<sub>x</sub> emissions, (2) higher isoprene emissions, (3) more detailed NMHC schemes, and possibly (4) improved parametrizations of processes such as photolysis, convection, and stratosphere-troposphere exchange. A series of sensitivity runs were performed with one model (C: FRSGC/UCI), using a variety of anthropogenic NO<sub>x</sub> emissions magnitudes and distributions (broadly representative of the pre-TAR simulations reported in TAR Table 4.12), in combination with low/high (220/500 TgC/yr) isoprene emissions. For this model, the change in tropospheric ozone production between that reported in TAR Table 4.12 and that calculated here (Table 5) is an increase of ~20%; with about half of this change due to an increase in NO<sub>x</sub> emissions (total NO<sub>x</sub> emissions rise by 21%), and the other half due to the increase in isoprene emissions. The year 2000 NO<sub>x</sub> emissions used in this study show an equatorward shift compared to data for the 1990s used in earlier studies, and this may also contribute to the increase in P. These sensitivity runs also show a reduction in ozone lifetime of 6% due to the changes in emissions, again with about half from NO<sub>x</sub> changes and half from isoprene changes. These largely explain the differences found in the ensemble mean ozone lifetimes between this study and TAR Table 4.12.

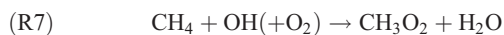
[41] The ozone burden has increased by 10% compared to TAR despite a 30% reduction in stratospheric input and a similar increase in dry deposition, and a decrease in the ozone lifetime. Models have clearly increased in chemical activity in the last 5 years.

[42] Table 6 shows absolute and percentage changes in ensemble mean O<sub>3</sub> budget terms between the various 2030 scenarios and 2000, together with standard deviations.



**Figure 8.** Constellations of ozone lifetime ( $\tau_{O_3}$ , days) plotted against ozone burden ( $B_{O_3}$ ,  $Tg(O_3)$ ) for individual models. Large solid (small open) circles are values for S1 (S2) for the 21 models that reported ozone budgets (Table 5). The letter refers to the model code (Table 2). Where models reported values for S3 and S4, lines extend to higher and lower burdens, respectively. The nine models that performed S5 and reported budgets are joined by dotted lines to S2 values, the scenario with the same emissions but without climate change. The dashed lines are isolines of overturning flux ( $F_{O_3}$ ), from 4000  $Tg(O_3) yr^{-1}$  (top left) to 8000  $Tg(O_3) yr^{-1}$  (bottom right). Ensemble mean results (Table 6) are plotted oversized in black; the inset shows ensemble mean results, using the same scales but offset axes, labeling the position of each scenario.

Changes in stratospheric influx of ozone are insignificant for all the scenarios with fixed meteorology, as would be expected; however with climate change, this term increases by 8%. The ozone lifetime slightly reduces under S2, while it increases under S3, and quite strongly reduces under S4. These changes occur due to changes in chemical and deposition loss rates. Since water vapor concentrations are fixed in these cases, chemical changes are mainly controlled by reactions (R4) and (R5), and reflect shifts in  $HO_x$  amounts and the  $OH/HO_2$  ratio [e.g., *Lelieveld et al.*, 2004]. This ratio is affected by emissions of  $NO$ , which tends to increase  $OH$ , via reaction (R1). Working in the opposite sense, emissions of  $CO$ ,  $CH_4$  and NMHCs, tend to increase  $HO_2$ , e.g., via the following reactions:



Deposition loss rates change as boundary layer ozone concentrations change; indeed, changes in the 3-D distribution of ozone also affect its overall mean tropospheric lifetime, as the lifetime shows substantial spatial variation.

[43] Climate change tends to further reduce the lifetime, mainly via increases in water vapor, promoting the major ozone loss reaction (R3). However, this is counteracted in some models by increased stratospheric influx, which

increases ozone concentrations in the upper troposphere, where its lifetime is longest.

[44] Figure 8 summarizes all the  $O_3$  budget results on a single diagram, plotting  $O_3$  lifetime against burden, with individual model's results for each scenario linked together. The dashed lines radiating from the origin represent lines of constant turnover flux (equation (3)). There is a clear correlation between models with a long ozone lifetime and high ozone burden, with S1 results congregating around a turnover flux of about 5000–6000  $Tg(O_3) yr^{-1}$ . There is also a clear tendency for the ozone lifetime to fall as  $NO_x$  emissions and the ozone burden increase (i.e., stepping through scenarios in the order S3–S1–S2–S4; see Figure 8, inset) for a given model. The two STOCHEM models (especially STOCHEM-HadAM3) and MOZECH have relatively short ozone lifetimes, while there is a larger grouping with relatively long lifetimes; the root cause of these differences are not obvious. Models with a larger spread between scenarios are more sensitive to changes in emissions; UM\_CAM is the most sensitive model. The impact of climate change on the ozone budgets for individual models is given in Table 7, and also shown by the dotted lines in Figure 8, which connect the S5 simulation (CLE emissions, 2030 climate) to the same emission scenario simulated with a year 2000 climate (S2). Eight out of nine models show a decrease in ozone lifetime due to climate change. Changes in burden are more variable, but on average show slight decreases (see Figure 8, inset).

**Table 7.** Background (S2) Tropospheric Water Vapor Burdens (Q) and Differences Between S5 and S2 (Differences due to Climate Change) in Surface Global Annual Mean Temperature ( $T_0$ ), Q, P, L, D,  $S_{\text{inf}}$ ,  $B_{\text{O}_3}$ ,  $\tau_{\text{O}_3}$ , and  $\tau_{\text{CH}_4}$  for the Subset of Models That Ran S5

Model Letter	Q, Pg-H <sub>2</sub> O	$\Delta T_0$ , K	$\Delta Q$ , Pg-H <sub>2</sub> O	$\Delta P$ , Tg(O <sub>3</sub> )yr <sup>-1</sup>	$\Delta L$ , Tg(O <sub>3</sub> )yr <sup>-1</sup>	$\Delta D$ , Tg(O <sub>3</sub> )yr <sup>-1</sup>	$\Delta S_{\text{inf}}$ , Tg(O <sub>3</sub> )yr <sup>-1</sup>	$\Delta B_{\text{O}_3}$ , Tg(O <sub>3</sub> )	$\Delta \tau_{\text{O}_3}$ , days	$\Delta \tau_{\text{CH}_4}$ , years
B	11.6	0.95	0.68	35	156	-19	101	-2.4	-0.65	-0.46
E	12.5	0.64	0.61	—	—	—	—	-7.1	—	-0.39
L	13.0	0.36	0.38	-5	45	-23	28	0.9	-0.03	-0.29
P	12.1	0.70	0.60	57	122	-13	50	0.0	-0.43	-0.44
Q	13.7	0.33	0.36	99	145	-12	33	1.5	-0.25	-0.28
R	13.4	0.31	0.30	0	30	-12	18	-3.4	-0.22	-0.20
T	13.2	0.66	0.62	122	146	-23	0	-8.7	-0.83	-0.52
U	11.9	0.85	0.95	5	45	-21	19	-6.6	-0.52	-0.64
Y	—	0.52	—	-57	-32	9	34	5.4	0.40	-0.06
Z	13.2 <sup>a</sup>	0.66 <sup>a</sup>	0.62 <sup>a</sup>	75	158	-5	76	-0.1	-0.75	-0.50

<sup>a</sup>Model Z (UM CAM) did not report temperatures or humidities, but it uses the same underlying GCM as model T (STOCHEM\_HadAM3), so these values are repeated.

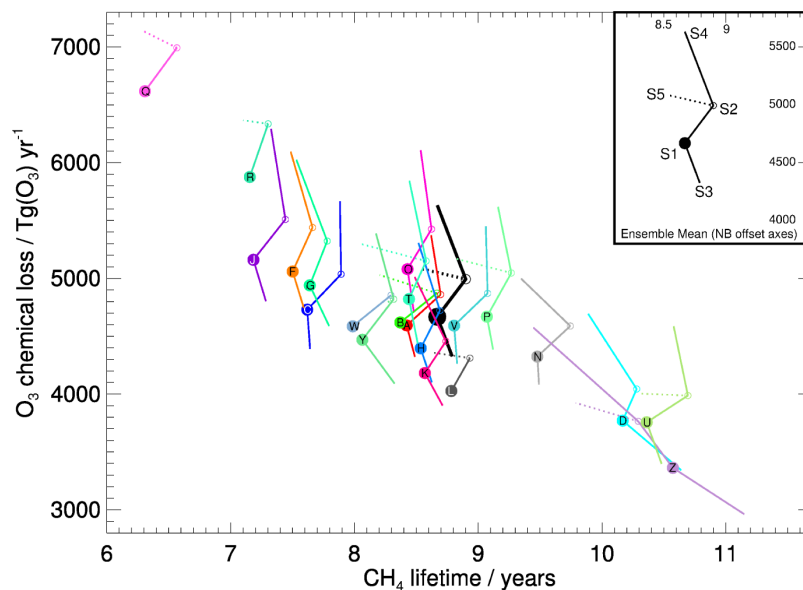
### 4.3. Methane Lifetimes

[45] Tables 5 and 6 also show whole atmosphere methane lifetimes ( $\tau_{\text{CH}_4}$ ). The major methane sink is oxidation by OH (R7). Modelers reported the mass flux through this reaction throughout their model domains; this was then masked above the chemical tropopause (using exactly the same method as above), to yield an annual mean tropospheric oxidation flux ( $F_{\text{CH}_4+\text{OH}}$ ). The other main loss processes for atmospheric methane are a soil sink ( $F_{\text{SOIL}}$ ) and a stratospheric sink ( $F_{\text{STRAT}}$ ). For these two sinks, we use fixed values of 30 Tg/yr and 40 Tg/yr, respectively, as recommended by Prather *et al.* [2001]. We converted specified methane mixing ratios (Table 3) to burdens ( $B_{\text{CH}_4}$ ) using an atmospheric mass of 5136 Eg. The methane lifetime is then given by

$$\tau_{\text{CH}_4} = B_{\text{CH}_4} / (F_{\text{CH}_4+\text{OH}} + F_{\text{SOIL}} + F_{\text{STRAT}}) \quad (4)$$

The ensemble mean ( $\pm 1$  SD) methane lifetime for S1 is  $8.7 \pm 1.3$  years (Table 5); this is in good agreement with the value of 8.4 years from TAR [Prather *et al.*, 2001, Table 4.3]. Figure 9 shows a clear relationship between the methane lifetime and ozone chemical loss rate (L); this can be understood in terms of reaction (R3), which is both the major ozone loss process, and the primary source of OH (the major methane loss process). In a similar way to Figure 8, Figure 9 summarizes results for all scenarios and all models, and also shows the ensemble mean values (Tables 5 and 6) in an inset. Most models show similar interscenario changes.

[46] The CLE scenario (S2) leads to a 3% increase in methane lifetime (compared to S1), whereas the MFR (S3) and A2 (S4) scenarios have insignificant effects. Under the 2030 climate change scenario (S5), the lifetime reduces by 4%; this reduction is a consistent finding across all nine models and reflects the higher levels of water vapor in a



**Figure 9.** Constellations of tropospheric ozone chemical loss rate ( $L$ , Tg(O<sub>3</sub>) yr<sup>-1</sup>) versus methane lifetime ( $\tau_{\text{CH}_4}$ , years) for individual models (S1 data in Table 5), following the same format as Figure 8. Inset shows the ensemble mean results (Table 6), labeling the position of each scenario.



warmer climate, and hence higher levels of OH and methane oxidation (reactions (R3) and (R7)). In addition, reaction (R7) is strongly temperature dependent, with warmer temperatures increasing the oxidation flux. Unlike ozone, there are no other significant climate feedback factors apparent in these simulations, although feedbacks associated with natural emissions (e.g., wetlands) are likely to be significant [e.g., *Gedney et al.*, 2004; *Shindell et al.*, 2004]; these are not included here.

[47] The spread in CH<sub>4</sub> lifetimes (and O<sub>3</sub> chemical loss rates) between models (Figure 9) is not fully understood, but part of the explanation is the underlying level of water vapor, which show some variation between models. Tropospheric water vapor burdens for eight of the GCMs are given in Table 7 and show that the wettest model (Q: MOZECH), also has the shortest CH<sub>4</sub> lifetime, and highest O<sub>3</sub> chemical loss rate (Table 5 and Figure 9). However, amongst the other models there is no clear correlation between high water vapor and these properties. A combination of factors, including both the HO<sub>x</sub> sources/sinks, and the OH/HO<sub>2</sub> partitioning must be important – more detailed HO<sub>x</sub> budgets are required to further understand these results.

#### 4.4. Ozone and Methane Radiative Forcings 2000–2030

[48] Ozone distributions from all models and all scenarios were inserted into an offline radiation code [*Edwards and Slingo*, 1996], and instantaneous short-wave and long-wave radiative fluxes at the tropopause were calculated. Each ozone distribution was overprinted with a fixed stratospheric ozone distribution above the chemical tropopause, eliminating any effect due to stratospheric ozone changes. Differencing the calculated fluxes between scenarios gives an instantaneous radiative forcing due to the change in tropospheric ozone. The code was set up as described by *Stevenson et al.* [1998], and included the effects of clouds but did not explicitly include stratospheric temperature adjustment. Previous studies with this set up have consistently found a 22% reduction in the instantaneous forcing compared to the equivalent forcing calculated following stratospheric adjustment; we apply this as a constant correction, to make the ozone forcings directly comparable to methane and CO<sub>2</sub> forcings from the same scenarios. Table 8 shows ensemble mean ( $\pm 1$ SD) tropospheric ozone forcings, together with the equivalent forcings for CH<sub>4</sub> and CO<sub>2</sub>. Methane forcings were calculated from the prescribed mixing ratios (Table 3), using the formula from IPCC TAR [*Ramaswamy et al.*, 2001, Table 6.2]; these values do not therefore represent output from this model exercise. Similarly, the CO<sub>2</sub> forcings are taken from IPCC TAR (Table II.3), for the A2 and B2 reference scenarios; CLE and MFR follow the B2 socioeconomic “story line” but impose additional measures to reduce some trace gas emissions (section 3.1); we assume these measures have no effect on CO<sub>2</sub> emissions.

[49] Following the CLE scenario, the combined methane and ozone forcing for the period 2000–2030 adds 23% to the B2 CO<sub>2</sub> forcing (Table 8). In stark contrast, the MFR scenario leads to a negative forcing, equivalent to a 6% reduction of the B2 CO<sub>2</sub> forcing. The higher emission A2 scenario adds 28% to the A2 CO<sub>2</sub> forcing (which is itself 31% larger than the B2 CO<sub>2</sub> value). By 2030, climate

**Table 8.** Radiative Forcings Between 2000 and the Three 2030 Scenarios and the Impact of Climate Change on the CLE Scenario in 2030 (S5–S2)<sup>a</sup>

	O <sub>3</sub> , mW m <sup>−2</sup>	CH <sub>4</sub> , mW m <sup>−2</sup>	CH <sub>4</sub> +O <sub>3</sub> , mW m <sup>−2</sup>	CO <sub>2</sub> , mW m <sup>−2</sup>
S2–S1	63 ± 15	116	180	780–810 (B2)
S3–S1	−45 ± 15	0	−50	780–810 (B2)
S4–S1	155 ± 37	141	300	1010–1080 (A2)
S5–S2	−3.4 ± 14	−26	−30	–

<sup>a</sup>Methane forcings are calculated from the prescribed global mixing ratios given in Table 3, using the formula of *Ramaswamy et al.* [2001, Table 6.2]; they do not therefore reflect the model responses. Carbon dioxide forcings from IPCC [2001, Table II.3] are given for comparison for the appropriate SRES scenario. Given the standard deviations of the ozone forcings, combined forcings are only quoted to the appropriate number of significant figures.

change is estimated to introduce a small negative feedback, about 4% of the B2 CO<sub>2</sub> forcing, mainly via methane. The effect of climate change on ozone, as discussed above, is rather uncertain, and may introduce either a small negative or positive feedback.

#### 4.5. Do Model Outliers Significantly Influence the Results?

[50] The model ensemble comprises a wide range of differently formulated models, and these models simulate present-day ozone to varying degrees of success (Figure 2). The models also exhibit a range of responses to future emissions and climate (Figures 8 and 9), and there are some clear outliers, in terms of their ozone budgets and methane lifetimes. To check if these outliers were significantly biasing the mean model, we selected a subset of models, using two criteria: (1) simulated O<sub>3</sub> has an RMSE less than 12.5 ppbv and (2) methane lifetime is within 1 standard deviation of the mean, i.e., within the range  $8.67 \pm 1.32$  years (Table 5). These criteria selected the subset of models shown in bold in Table 5, which also shows the mean O<sub>3</sub> budget terms and methane lifetime based on this subset of models. The mean values are very similar to the mean of the whole ensemble, indicating that the outliers have little influence on the mean values for the present-day simulation. As would be expected, using this subset reduces the standard deviations of most terms. We also checked if this subset of models produced different results for the various 2030 scenarios, and found that there was no significant influence on the results. We conclude that the outliers have little influence on the mean response, and that use of the entire ensemble represents a robust method for assessing future levels of ozone and quantitatively assessing uncertainties.

### 5. Summary and Conclusions

[51] This study synthesizes results from a wide range of models (up to 26 different models were used), which collectively represent the best available method for simulating tropospheric composition. Using a model ensemble approach improves the robustness of results, and allows quantification of uncertainties. By considering a range of emission scenarios covering the next thirty years, we have produced quantitative projections of the likely outcomes

from different future regulatory options that are currently open to the world.

[52] Comparison of the ensemble mean model year 2000 simulation with ozonesonde observations (Figure 2) shows good agreement. Column  $\text{NO}_2$  distributions from the models also compare well with satellite observations [van Noije *et al.*, 2006]. This suggests that the emissions' specifications and the models' simulation of ozone are consistent with the real atmosphere. The mean model's methane lifetime of  $8.7 \pm 1.3$  years (Table 5) is also in agreement with observationally derived estimates [e.g., Lawrence *et al.*, 2001; Krol and Lelieveld, 2003; Bloss *et al.*, 2005; Bousquet *et al.*, 2005; Prinn *et al.*, 2005], indicating that the mean model realistically simulates the global distribution of OH.

[53] The mean model's tropospheric ozone budget for the year 2000 (Table 5) shows distinct differences compared to the budgets reported in IPCC TAR [Prather *et al.*, 2001, Table 4.12]: chemical production is higher by nearly 50%, chemical destruction and dry deposition are larger by 30%, while stratospheric input is 30% less. The mean ozone lifetime is 10% less, while the ozone burden is 10% larger. The reasons for these changes are not immediately obvious, but probably partially relate to the higher total  $\text{NO}_x$  emissions used here compared to earlier studies; also isoprene emissions are somewhat higher; and NMHC schemes have developed in sophistication over the last 5 years. The sampled models are also different, although a significant subset of the TAR models (and their descendents) is also represented here. The higher value of the simulated present-day ozone burden may lead to a larger estimate of the preindustrial to present-day radiative forcing from tropospheric ozone (although this is not the focus of this study).

[54] Three emission scenarios for the year 2030 were simulated, broadly representing "likely" (S2, CLE), "optimistic" (S3, MFR) and "pessimistic" (S4, A2) future situations. These span global  $\text{NO}_x$  emission changes, compared to year 2000 (S1), of +12%, -27% and +55% (Table 3) respectively, and yield changes in tropospheric  $\text{O}_3$  burden of +6%, -5%, and +15% (Table 6 and Figures 3–6), and net  $\text{O}_3 + \text{CH}_4$  radiative forcings for 2000 to 2030 of +180, -50 and +300  $\text{mW m}^{-2}$  (Table 8). Simulated changes and forcings have typical intermodel uncertainties ( $\pm 1$  SD) of 20–35%. These results illustrate the sensitivity of ozone to anthropogenic emissions changes, and hence the degree of human control over this key gas.

[55] A further influence on future tropospheric composition considered here is climate change. By 2030, surface warming on the order of 0.7 K is predicted [IPCC, 2001]. Ten of the models performed a climate change simulation (S5), repeating S2 but changing the underlying climate to one appropriate for 2030. This ensemble produced quite divergent results (Table 7) but identified two major feedbacks on ozone that compete for dominance. The first mechanism, apparent to some extent in all the models, involves the increase in water vapor that accompanies warming, which promotes ozone destruction, via reaction (R3). This feedback is strongest in the tropical lower troposphere, particularly over the oceans (Figure 7), and is diagnosed as an increase in chemical loss of ozone (Tables 6 and 7). The second mechanism, apparent in all but one of the models, is an increase in the stratospheric influx of ozone (Tables 6 and 7), tending to increase upper tropo-

spheric ozone, particularly in the Northern Hemisphere. The mean impact of climate change on ozone burden and radiative forcing is a small, but insignificant decrease (Tables 6 and 8).

[56] Climate change has a more consistent impact on methane, increasing its oxidation through both increased levels of OH (also via reaction (R3)), and higher temperatures, which increase the rate coefficient for reaction (R7). The ensemble mean methane lifetime reduces by 4% (Table 6), lowering methane concentrations, and hence radiative forcing (Table 8).

[57] Intermodel standard deviations provide a measure of uncertainty in our ability to simulate ozone. Model simulations of the present-day ozone distribution show the best agreement in the Northern midlatitude midtroposphere (Figure 1c:  $\pm 12$ –14%); whereas the models are least consistent throughout the Antarctic troposphere, in the upper troposphere of the Southern Hemisphere and tropics, and in the Arctic lower troposphere ( $> \pm 30\%$ ). Models also show less agreement in the tropics compared to midlatitudes; this probably reflects the larger uncertainties associated with tropical processes such as deep convection, isoprene emissions and chemistry, lightning  $\text{NO}_x$ , and biomass burning emissions. Northern midlatitudes are the most intensively observed atmospheric region, so we might expect model uncertainties to be lowest here.

[58] We find that model outliers have little influence on the model mean, but, as would be expected, increase intermodel standard deviations, and hence our estimates of model uncertainty. Use of the entire model ensemble gives the best estimate of the mean model response and its uncertainty.

[59] To conclude, this study has shown the extent of anthropogenic control over tropospheric ozone and the oxidizing capacity of the atmosphere, and also the likely effects of climate change over the time period 2000–2030. There are clear benefits to following a trajectory of global emissions reductions, in terms of reducing the radiative forcing of climate, and also for improving air quality (Ellingsen *et al.*, in preparation, 2006) and reducing the environmental impacts from nitrogen and sulphur deposition (Dentener *et al.*, submitted manuscript, 2005). By contrast, the higher emission scenario provides a stark warning of the likely environmental consequences of failing to adhere to current legislation.

[60] All modeling studies have inherent uncertainties; this study uses a large model ensemble approach to reduce and quantify these. In order to reduce these uncertainties, future work will need to focus on the most poorly understood model processes and variables; these include: convection, water vapor, biogenic hydrocarbon emissions and chemistry, natural  $\text{NO}_x$  emissions, stratosphere-troposphere exchange, and biomass burning.

## Appendix A

[61] Table A1 provides a brief summary of information for each of the models, comprising names of the authors responsible for contributing results; model resolution; driving meteorology, including whether the model is a CTM driven by meteorological analyses, or if it is driven by GCM output; details of the convection, advection and tropo-

Table A1. List of Models in Detail

Model	Institute	Contact Author	Resolution Longitude/Latitude/Levels, Top Level	Underlying Meteorology	Advection Scheme	Convection Scheme	Tropospheric Chemistry	Stratospheric Chemistry	GCM Coupling Chemistry to Radiation	References
A. CHASER_CTM	FRCGC/JAMSTEC	Kengo Sudo	2.8°/2.8°/L32, 3 hPa	CTM: ECMWF	<i>Lin and Rood</i> [1996]	Prognostic Anrakawa-Schubert scheme in CCSR/NIES GCM	53 species Interactive SO <sub>2</sub> aerosol	O <sub>3</sub> relaxed above 50 hPa to observations		<i>Sudo et al.</i> [2002a, 2002b, 2003]
B. CHASER_GCM	FRCGC/JAMSTEC	Kengo Sudo	2.8°/2.8°/L32, 3 hPa	GCM: CCSR/NIES	<i>Lin and Rood</i> [1996]	Prognostic Anrakawa-Schubert scheme in CCSR/NIES GCM	53 species Interactive SO <sub>2</sub> aerosol	O <sub>3</sub> relaxed above 50 hPa to observations	Coupled (O <sub>3</sub> , H <sub>2</sub> O, N <sub>2</sub> O, CH <sub>4</sub> , CFCs)	<i>Sudo et al.</i> [2002a, 2002b, 2003]
C. FRSGC/UCI	FRCGC/JAMSTEC	Oliver Wild	2.8°/2.8°/L37, 10 hPa	CTM: ECMWF-JFS pieced-forecast data for 2000	<i>Prather</i> [1986]	Mass fluxes taken from ECMWF-JFS fields, based on <i>Tiedtke</i> [1989]	35 species (27 transported), using ASAD [Carver et al., 1997]	LINOZ[McLinden et al., 2000]		<i>Wild and Prather</i> [2000], <i>Wild et al.</i> [2003]
D. GEOS-CHEM	LMCA-EPFL	Isabelle Bey and Jerome Drevet	5°/4°/L30, 0.01 hPa	CTM: GEOS winds NASA GMAO	<i>Lin and Rood</i> [1996]	Mass fluxes taken from GEOS-3 assimilated fields	31 tracers (24 for O <sub>3</sub> , 7 for SO <sub>2</sub> -NH <sub>3</sub> -NO <sub>y</sub> aerosols, interactive).	SYNOZ: O <sub>3</sub> production rate at 70 hPa: 500 Tg/yr aerosols, [McLinden et al., 2000]		<i>Bey et al.</i> [2001], <i>Martin et al.</i> [2003], <i>Park et al.</i> [2004]
E. GISS	NASA GISS/Columbia University, New York	Nadine Bell and Drew T. Shindell	5°/4°/L23, 0.01 hPa	GCM: ModelE GISS	<i>Prather</i> [1986]	<i>DeGuent et al.</i> [1996]	35 species 20 tracersInteractive SO <sub>2</sub> aerosol	O <sub>3</sub> climatology based on satellite and sonde observations. NO <sub>y</sub> set to prescribed NO <sub>x</sub> /O <sub>3</sub> ratios	Decoupled	<i>Shindell et al.</i> [2003, 2005], <i>Bell et al.</i> [2005], <i>Schmidt et al.</i> [2006]
F. GMI/CCM3	NASA Global Modeling Initiative	Jose M. Rodriguez and Susan Strahan	5°/4°/L52, 0.006 hPa	GCM: NCAR MACCM3	<i>Lin and Rood</i> [1996]	Mass fluxes taken from CCM3 deep convection	85 species Off-line SO <sub>x</sub> aerosol	O <sub>3</sub> influx from SYNOZ: 550 Tg/yr		<i>Rotman et al.</i> [2001], <i>Wild et al.</i> [2001], <i>Bey et al.</i> [2001]
G. GMI/DAO	NASA Global Modeling Initiative	Jose M. Rodriguez and Susan Strahan	5°/4°/L46, 0.048 hPa	CTM: GEOS-2-DAS Assimilated fields for Mar 1997–Feb 1998.	<i>Lin and Rood</i> [1996]	Mass fluxes taken from GEOS-2 assimilated fields	85 species Off-line SO <sub>x</sub> aerosol	O <sub>3</sub> influx from SYNOZ: 550 Tg/yr		<i>Rotman et al.</i> [2001], <i>Wild et al.</i> [2001], <i>Bey et al.</i> [2001]
H. GMI/GISS	NASA Global Modeling Initiative	Jose M. Rodriguez and Susan Strahan	5°/4°/L23 0.017 hPa	GCM: GISS-2 <sup>o</sup>	<i>Lin and Rood</i> [1996]	Mass fluxes for deep/shallow convection from GISS-II <sup>o</sup> , as corrected by M. J. Prather (private communication, 2004)	85 species Off-line SO <sub>x</sub> aerosol	O <sub>3</sub> influx from SYNOZ: 550 Tg/yr		<i>Rotman et al.</i> [2001], <i>Wild et al.</i> [2001], <i>Bey et al.</i> [2001]
I. IASB	IASB/Belgium	Jean-Francois Müller	5°/5°/L25, 50 hPa	CTM: Monthly means from ECMWF reanalyses (1993–2001 ERA40)	<i>Smolarkiewicz and Rasch</i> [1991]	<i>Costen et al.</i> [1998]; cumulonimbus distribution taken from ISCCP	60 species (40 transported) Interactive SO <sub>x</sub> aerosol	Zonal mean O <sub>3</sub> prescribed above 98 hPa, zonal mean HNO <sub>3</sub> prescribed above 50 hPa		<i>Miller and Brasseur</i> [1995], <i>Miller and Stavrakou</i> [2005]
J. LLNL-IMPACT	Lawrence Livermore National Laboratory	Cynthia S. Atherton and Daniel J. Bergmann	5°/4°/L26 2 hPa	GCM: CAM3	<i>Lin and Rood</i> [1996]	<i>Zhang and McFarlane</i> [1995] (deep) and <i>Hack</i> [1994] (shallow)	100 species Including Cl <sub>x</sub> , Br <sub>y</sub>	Full stratospheric chemistry, including Cl <sub>x</sub> , Br <sub>y</sub>	Decoupled	<i>Rotman et al.</i> [2004]
K. LMDz/INCA-CTM	LSCE	Didier Hauglustaine and Sophie Szopa	3.75°/2.5°/L19, 3hPa	CTM:nudged to ECMWF/ERA15-ERA40-OD	<i>van Leer</i> [1977]	<i>Tiedtke</i> [1989]	85 species	Stratospheric O <sub>3</sub> nudged toward climatology above 380 K		<i>Sadourny and Laval</i> [1984], <i>Hauglustaine et al.</i> [2004]
L. LMDz/INCA-GCM	LSCE	Didier Hauglustaine and Sophie Szopa	3.75°/2.5°/L19 3hPa	GCM: LMDz	<i>van Leer</i> [1977]	<i>Tiedtke</i> [1989]	85 species	Stratospheric O <sub>3</sub> nudged toward climatology above 380 K	Decoupled	<i>Sadourny and Laval</i> [1984], <i>Hauglustaine et al.</i> [2004]



Table A1. (continued)

Model	Institute	Contact Author	Resolution Longitude/Latitude/Levels, Top Level	Underlying Meteorology	Advection Scheme	Convection Scheme	Tropospheric Chemistry	Stratospheric Chemistry	GCM Coupling Chemistry to Radiation	References
M. MATCH-MPIC/ ECMWF	Max Planck Institute for Chemistry/NCAR	Tim Butler and Mark Lawrence	5.6°/5.6°/L60, 0.1 hPa	CTM:ECMWF reanalysis	<i>Rasch and Lawrence</i> [1998]	<i>Zhang and McFarlane</i> [1995] (deep) and <i>Hack</i> [1994] (shallow)	60 species	Zonal mean O <sub>3</sub> climatology above 30 hPa above the tropopause; NO <sub>x</sub> set to prescribed NO <sub>x</sub> /O <sub>3</sub> ratios		<i>von Kuhlmann et al.</i> [2003a, 2003b], <i>Lawrence et al.</i> [1999], <i>Rasch et al.</i> [1997]
N. MATCH-MPIC/ NCEP	Max Planck Institute for Chemistry/NCAR	Tim Butler and Mark Lawrence	5.6°/5.6°/L60, 0.1 hPa	CTM:NCEP/NCAR reanalysis	<i>Rasch and Lawrence</i> [1998]	<i>Zhang and McFarlane</i> [1995] (deep) and <i>Hack</i> [1994] (shallow)	60 species	Zonal mean O <sub>3</sub> climatology above 30 hPa above the tropopause; NO <sub>x</sub> set to prescribed NO <sub>x</sub> /O <sub>3</sub> ratios		<i>von Kuhlmann et al.</i> [2003a, 2003b] <i>Lawrence et al.</i> [1999] <i>Rasch et al.</i> [1997]
O. MOZZ-GFDL	GFDL	Arlene Fiore and Larry Horowitz	1.9°/1.9°/L28, 0.7 hPa	CTM: NCEP reanalysis	<i>Lin and Rood</i> [1996]	<i>Zhang and McFarlane</i> [1995] (deep) and <i>Hack</i> [1994] (shallow)	74 species Interactive SO <sub>x</sub> , BC aerosols	Overwrite O <sub>3</sub> above 14 hPa; between 14 hPa and tropopause relax to climatology (10 day time constant). Relaxed to <i>Logan</i> [1999] above tropopause; above 100 hPa	Decoupled	<i>Brasseur et al.</i> [1998], <i>Hauglustaine et al.</i> [1998], <i>Horowitz et al.</i> [2003]
P. MOZART4	NCAR	Jean Francois Lamarque	2.8°/2.8°/L26, 4 hPa	GCM:CCSM3	<i>Lin and Rood</i> [1996]	<i>Zhang and McFarlane</i> [1995] (deep) and <i>Hack</i> [1994] (shallow)	96 species Aerosols SO <sub>x</sub> , NH <sub>3</sub> , NO <sub>x</sub> , BCOC, seasalt, SOA.	Relaxed to climatology (10 day time constant). Relaxed to stratospheric O <sub>3</sub> , NO <sub>x</sub> , and HNO <sub>3</sub> down to 200 hPa in extra-tropics and 100 hPa in tropics (10 day time constant)	Decoupled	<i>Horowitz et al.</i> [2003], <i>Tie et al.</i> [2005], <i>Lamarque et al.</i> [2003a, 2003b], Enmons et al. (manuscript in preparation, 2006) S. Rast et al. (manuscript in preparation, 2006), ECHAM5 described by <i>Röckner et al.</i> [2003], MOZART-2 described by <i>Horowitz et al.</i> [2003]
Q. MOZECH	MPI Meteorology	Martin G. Schultz and Sebastian Rast	1.9°/1.9°/L31, 10 hPa	GCM: ECHAM5	<i>Lin and Rood</i> [1996]	<i>Tiedtke</i> [1989] with modifications after <i>Nordeng</i> [1994]	63 species SO <sub>x</sub> climatology	Relaxed to stratospheric O <sub>3</sub> , NO <sub>x</sub> , and HNO <sub>3</sub> down to 200 hPa in extra-tropics and 100 hPa in tropics (10 day time constant)	Decoupled	Rast et al. (manuscript in preparation), ECHAM5 described by <i>Röckner et al.</i> [2003] MOZART-2 described by <i>Horowitz et al.</i> [2003]
R. MOZECH2	MPI Meteorology	Martin G. Schultz and Sebastian Rast	2.9°/2.9°/L31, 10 hPa	GCM: ECHAM5	<i>Lin and Rood</i> [1996]	<i>Tiedtke</i> [1989] with modifications after <i>Nordeng</i> [1994]	As MOZECH, but: Lightning NOx reduced to 3.7 Tg N yr <sup>-1</sup> H <sub>2</sub> O vapor > 700 hPa reduced by 10% in chemistry/surface albedo from <i>Laegle et al.</i> [2005]	Relaxed to stratospheric O <sub>3</sub> , NO <sub>x</sub> , and HNO <sub>3</sub> down to 200 hPa in extra-tropics and 100 hPa in tropics (10 day time constant)	Decoupled	
S. p-TOMCAT	University of Cambridge, UK	Nick Savage and John A. Pyle	2.8°/2.8°/L31, 10 hPa	CTM:ECMWF Operational analysis data	<i>Prather</i> [1986]	<i>Tiedtke</i> [1989]	37 species (24 transported) No aerosol	O <sub>3</sub> fixed above 10 hPa based on 2D model		<i>Law et al.</i> [1998, 2000]
T. STOCHEM- HadAM3	University of Edinburgh	David Stevenson and Ruth Doherty	5°/5°/L9 100 hPa	GCM:HadAM3 vn4.5	<i>Collins et al.</i> [1997]	<i>Collins et al.</i> [2002]	70 species SO <sub>x</sub> -NO <sub>x</sub> -NH <sub>3</sub> aerosols; interactive	Prescribed O <sub>3</sub> concentration gradient at 100 hPa	Decoupled	<i>Collins et al.</i> [1997], <i>Stevenson et al.</i> [2004]
U. STOCHEM- HadGEM	UK Met Office	Michael Sanderson and Bill Collins	3.75°/2.5°/L20, 40 km	GCM:HadGEM	<i>Collins et al.</i> [1997]	<i>Collins et al.</i> [2002]	70 species SO <sub>x</sub> -NO <sub>x</sub> -NH <sub>3</sub> aerosols; interactive	Relaxed toward SPARC O <sub>3</sub> climatology above tropopause	Decoupled	<i>Collins et al.</i> [1997, 2003]

Table A1. (continued)

Model	Institute	Contact Author	Resolution Longitude/Latitude/Levels, Top Level	Underlying Meteorology	Advection Scheme	Convection Scheme	Tropospheric Chemistry	Stratospheric Chemistry	GCM Coupling Chemistry to Radiation	References
V. TM4	KNMI	Twan van Noije	3°/2°/L25, 0.48 hPa	CTM: ECMWF 3-6-h operational forecasts (2000)	Russel and Lerner [1981]	Tiedtke [1989]	37 species (22 transported) SO <sub>2</sub> -NO <sub>y</sub> -NH <sub>x</sub> aerosols, interactive	O <sub>3</sub> nudged toward climatology above 123 hPa; except 30N–30S, above 60 hPa		Dentener <i>et al.</i> [2003], van Noije <i>et al.</i> [2004]
W. TM5	JRC	Frank Dentener and Maarten Krol	6°/4°/L25, 0.48 hPa (1°/1° Europe, North America, and Asia)	CTM: ECMWF 3-6-h operational forecasts (2000)	Russel and Lerner [1981]	Tiedtke [1989]	37 species (22 transported) SO <sub>2</sub> -NO <sub>y</sub> -NH <sub>x</sub> aerosols, interactive	O <sub>3</sub> nudged toward climatology above 50 hPa		Dentener <i>et al.</i> [2003], Krol <i>et al.</i> [2005]
X. UIO_CTM2	University of Oslo	Kjerstin Ellingsen and Michael Gauss	2.8°/2.8°/L40, 10 hPa	CTM: ECMWF-IFS forecast data	Prather [1986]	Tiedtke [1989]	58 species	O <sub>3</sub> , HNO <sub>3</sub> , and NO <sub>x</sub> from OsloCTM2 model run with stratospheric chemistry		Sindet [1997]
Y. ULAQ	Università L'Aquila	Veronica Montanaro and Giovanni Pitari	22.5°/10°/L26, 0.04 hPa	GCM: ULAQ-GCM	Eulerian flux form	Pitari <i>et al.</i> [2002] following Müller and Brasseur [1995]	Includes tropospheric aerosols	Detailed stratospheric chemistry scheme, including stratospheric aerosols	Coupled (CO <sub>2</sub> , H <sub>2</sub> O, CH <sub>4</sub> , O <sub>3</sub> , N <sub>2</sub> O, CFCs, HCFCs, aerosols)	Pitari <i>et al.</i> [2002]
Z. UM_CAM	University of Cambridge	Guang Zeng and John Pyle	3.75°/2.5°/L19, 4.6 hPa	GCM: HadAM3 vln4.5	Leonard <i>et al.</i> [1995]	Rowntree [1990]	60 species (36 transported) No aerosols.	O <sub>3</sub> and NO <sub>y</sub> prescribed above 30 hPa (Tropospheric chemistry operates below 30 hPa)	Decoupled	Zeng and Pyle [2003, 2005]

spheric/stratospheric chemical schemes; details of any coupling between chemistry and radiation schemes in the GCM-based models; and finally a list of references giving a more detailed model description.

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