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Future tropospheric ozone simulated with a climate-chemistry-biosphere model

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[1] A climate-chemistry model and a biogenic emission model are used to investigate the relative impact of anthropogenic and biogenic emissions of ozone precursors and global warming on the evolution of ozone in 2100. A warmer and wetter climate leads to enhanced ozone photochemical destruction in the lower troposphere, a more intense Brewer-Dobson circulation in the stratosphere and a lightning NO_x emission increased from 5 to 7.5 $\text{Tg(N)}/\text{yr}$. Over Europe and the eastern US climate change locally causes surface ozone to increase because of enhanced PAN thermal decomposition and more stagnant meteorological conditions. The global and annual mean OH concentration remains quite stable and the methane lifetime is unchanged in the future. Increased biogenic emissions contribute by 30–50% to surface ozone summer formation in northern continental regions. The feedback of climate change and of biogenic emissions increases the 2000 to 2100 tropospheric ozone radiative forcing by 12%, up to a global mean value of 0.58 Wm^{-2} . **Citation:** Hauglustaine, D. A., J. Lathière, S. Szopa, and G. A. Folberth (2005), Future tropospheric ozone simulated with a climate-chemistry-biosphere model, *Geophys. Res. Lett.*, 32, L24807, doi:10.1029/2005GL024031.

1. Introduction

[2] Ozone (O_3) is produced in the troposphere from the oxidation of carbon monoxide (CO) and volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x) and sunlight. Several modeling studies suggest that ozone could increase during the 21st century as a direct consequence of enhanced anthropogenic emissions of these precursors [Hauglustaine and Brasseur, 2001; Prather et al., 2001]. The projection of future tropospheric ozone needs to account for several feedbacks which could significantly affect tropospheric chemistry. The physical climate change and in particular the change in temperature and water vapor have been shown to significantly alter these projections [Brasseur et al., 1998; Stevenson et al., 2000]. Input from the stratosphere is also a significant source of ozone in the troposphere and changes in the atmospheric circulation under an evolving climate has also been shown to impact tropospheric ozone in the future [Zeng and Pyle, 2003; Collins et al., 2003]. In addition to that, the biosphere represents a major source

of ozone precursors such as VOCs from vegetation and NO_x from soils. Future climate changes could significantly affect these emissions and hence perturb tropospheric chemistry [Sanderson et al., 2003; Lathière et al., 2005]. The uncertainties on future ozone levels arise from the relative importance of these feedbacks between different models. In this study, we apply a global three-dimensional climate chemistry model and a biogenic emission model to investigate the relative impact of anthropogenic emissions, future natural emissions of VOCs and NO_x and physical climate change on the distribution and budget of tropospheric ozone and global OH concentration in the year 2100.

2. The Chemistry-Climate Model

[3] LMDz (Laboratoire de Météorologie Dynamique, zoom) is a grid point General Circulation Model (GCM) used in this study with a horizontal resolution of 3.8 degrees in longitude and 2.5 degrees in latitude and has 19 vertical σ -p levels extending from the surface up to 3 hPa. LMDz has been coupled on-line to INCA (Interactive Chemistry and Aerosols) [Hauglustaine et al., 2004]. The version of INCA used in this study includes 85 chemical species and 264 chemical reactions, and simulates tropospheric chemistry, emissions, and deposition of primary tropospheric trace species including non-methane hydrocarbons. The ORCHIDEE (Organizing Carbon and Hydrology in Dynamic Ecosystems) dynamical vegetation model [Krinner et al., 2005] has been used to calculate biogenic surface fluxes of isoprene, terpenes, acetone, and methanol and NO soil emissions as described by Lathière et al. [2005]. Lightning NO_x emissions are parameterized according to Price and Rind [1992] as described by Jourdain and Hauglustaine [2001]. For both present and future simulations we assume a vertical distribution based on Pickering et al. [1998].

3. Performed Simulations

[4] We performed a set of four simulations. Experiment “2000”: in this experiment, present-day climate conditions and surface emissions of precursors are considered (see auxiliary material¹). Total surface emissions are provided in Table 1. Experiment “2100A”: climate

Table 1. Emission Scenarios, Calculated Ozone Budget Terms and Adjusted Net Radiative Forcing^a

	2000	2100A	2100AB	2100ABC
NO _x	45.4	122.6	124.4	124.4
CO	1199.9	2648.2	2648.2	2648.2
CH ₄	562.8	1128.6	1128.6	1128.6
Anthropogenic VOCs	175.4	322.8	322.8	322.8
Isoprene	402.0	402.0	637.9	637.9
Terpenes	130.8	130.8	265.3	265.3
Other BVOCs	163.2	163.2	287.8	287.8
O ₃ production	4758.6	8586.0	9366.3	9750.8
O ₃ destruction	4157.6	7083.5	7746.9	8439.4
O ₃ net chemistry	601.0	1502.5	1619.4	1311.4
O ₃ influx	677.7	472.0	496.4	671.4
O ₃ dry deposition	1278.8	1974.5	2115.8	1982.8
O ₃ burden	292.4	427.4	444.7	438.8
O ₃ lifetime	25.7	22.0	21.0	19.0
OH	10.4	9.0	8.7	9.8
CH ₄ lifetime	10.2	11.7	11.9	10.2
O ₃ forcing	–	0.521	0.581	0.585

^aNO_x emissions (Tg(N)/yr) include surface, aircraft, and lightning contributions. CO and CH₄ emissions in Tg/yr. Isoprene, terpenes, BVOC and VOC emissions in Tg(C)/yr. Other BVOCs: methanol and acetone. O₃ production, destruction, net chemistry (production-destruction), dry deposition and stratospheric influx across 200 hPa in Tg/yr. O₃ burden in Tg. O₃ lifetime in days. OH in 10⁵ molec. cm⁻³. Methane lifetime in years. O₃ net radiative forcing at the tropopause in W/m².

conditions and future biogenic emissions are identical to exp. 2000. Anthropogenic emissions of ozone precursors for the year 2100 are based on the IPCC Special Report on Emission Scenarios (SRES) scenario A2 [Nakićenović *et al.*, 2000]. A similar scenario was used by Stevenson *et al.* [2000], Zeng and Pyle [2003], and Prather *et al.* [2001] and is applied again in our study. Experiment “2100AB”: present-day climate conditions are used as for the two previous experiments but both future anthropogenic and biogenic emissions of precursors are considered. The future seasonally varying emissions of Biogenic Volatile Organic Compounds (BVOCs) and NO from soils are prescribed based on simulations with the ORCHIDEE vegetation model as described by Lathière *et al.* [2005]. As a consequence of climate and vegetation changes, the NO soil emissions increase from 8.6 Tg(N)/yr for present-day conditions to 10.5 Tg(N)/yr, and isoprene and terpene emissions increase by 59% and 102%, respectively (Table 1). The total BVOC emissions significantly increase from 696 Tg(C)/yr for the present to 1191 Tg(C)/yr in the future. Experiment “2100ABC”: the future anthropogenic and biogenic emissions are considered for this simulation as well as future climate conditions. The changes in sea-surface temperature and sea-ice coverage in 2100 are taken from a simulation of the IPSL coupled model for scenario A2 [Dufresne *et al.*, 2002]. The global and decadal mean surface temperature increases by 2.45 K. This value is close to the 3.0 K average (with a range of 1.3 to 4.5 K) calculated by Cubasch *et al.* [2001] for this scenario. In all experiments, stratospheric ozone concentrations are relaxed toward climatologies above 150 hPa. This climatology is deliberately kept fixed at present-day values in all simulations in order to isolate the effect of tropospheric chemistry and climate change to the change in stratospheric chemistry. All simulations are run for a period of

5 years and the last two years of the simulations are used for analysis.

4. Results

[5] Figure 1 shows the calculated changes in zonal and annual mean tropospheric ozone. Figure 1a illustrates the difference between ozone calculated for experiment 2000 and experiments 2100ABC and 2100AB. For both future simulations, ozone increases throughout the whole troposphere. A maximum increase reaching 70% and 65% is calculated in the upper troposphere for exp. 2100ABC and 2100AB, respectively. In the lower troposphere, ozone increases by 60% in the tropics and by 40% at higher latitudes in exp. 2100ABC. A more pronounced increase reaching 70% is calculated for exp. 2100AB in the tropical lower troposphere. These results are generally consistent with the findings of Stevenson *et al.* [2000] and Zeng and Pyle [2003]. Table 1 provides the calculated ozone budget terms and global OH concentration for the different scenarios. The increase in anthropogenic and biogenic emissions in 2100 is responsible for an enhanced ozone net chemical production by more than a factor of two. The tropospheric ozone burden increases from 292 Tg for the present to 427 and 445 Tg for experiments 2100A and 2100AB, respectively. Anthropogenic and biogenic emissions reduce global OH by 16% and consequently the methane lifetime increases from 10.2 yr in 2000 to 11.9 yr in 2100 (exp. 2100AB). As a consequence of the enhanced upward transport of ozone across the 200 hPa isosurface in the tropics the net ozone influx into the defined tropospheric domain (i.e., below the 200 hPa model level) is reduced from 678 Tg/yr for present day to 472 and 496 Tg for exp. 2100A and 2100AB, respectively.

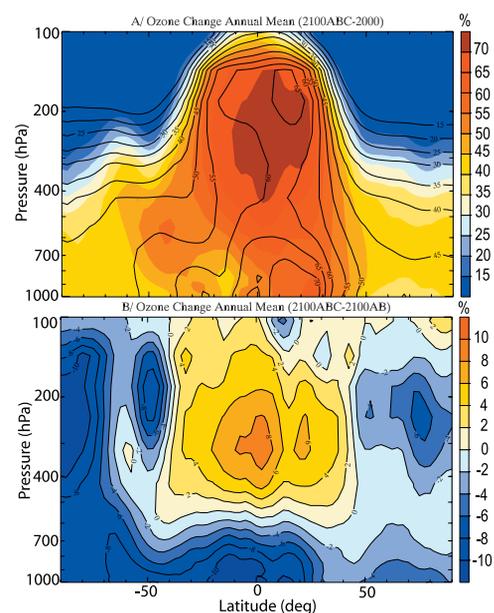


Figure 1. (a) Annual and zonal mean ozone change in 2100 (simulation 2100ABC–2000). The isolines show the ozone change calculated for 2100AB–2000. (b) Impact of climate change on ozone (simulation 2100ABC–2100AB). Note the different color scales. Units: %.

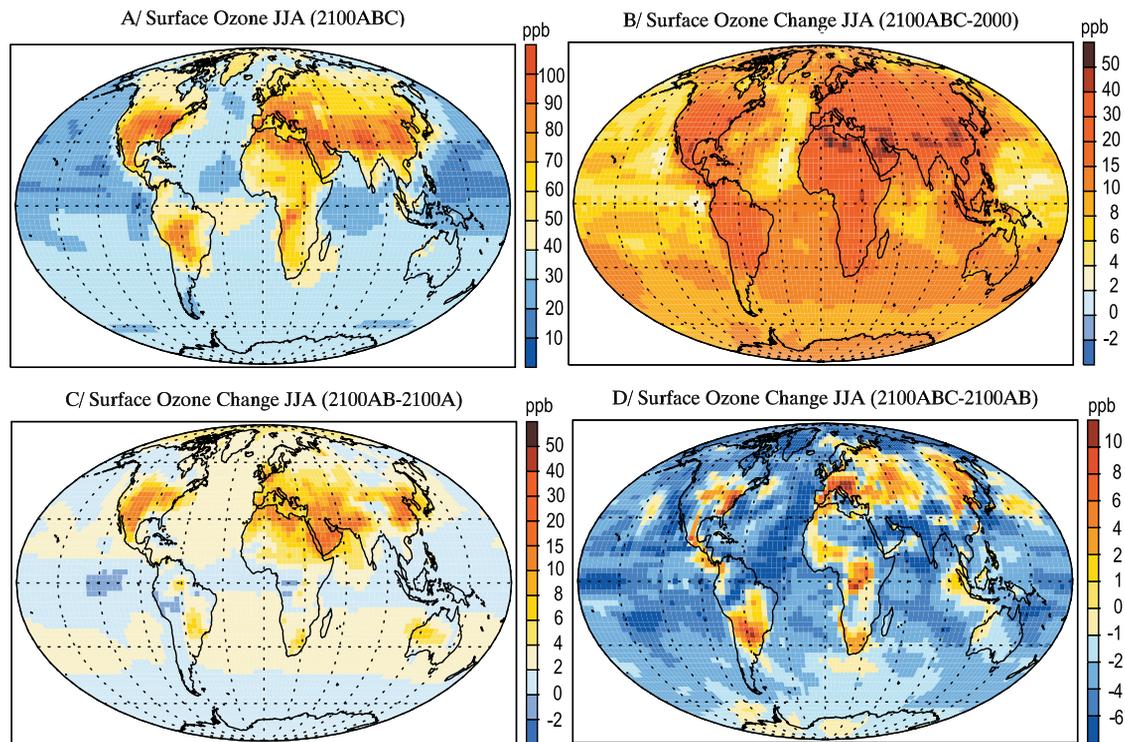


Figure 2. (a) Surface ozone calculated in 2100 for June–July–August (simulation 2100ABC). (b) Change in surface ozone in JJA 2100 (simulation 2100ABC–2000). (c) Change in surface ozone associated with biogenic emissions only (simulation 2100AB–2100A). (d) Impact of climate change on surface ozone (simulation 2100ABC–2100AB). Note the different color scales for Figures 2a and 2d. Units: ppb.

[6] Figure 1b illustrates the impact of climate change on ozone (exp. 2100ABC minus 2100 AB). In the lower troposphere (below about 500 hPa), the increase in water vapor reduces ozone by up to 10% in the tropics through the $\text{H}_2\text{O} + \text{O}(^1\text{D})$ reaction [Stevenson *et al.*, 2000]. The warmer and wetter climate causes global OH to increase from 8.7 to 9.8×10^5 molec. cm^{-3} (Table 1). Interestingly, the competing effects of emissions and climate change maintain global OH close to its present-day value in experiment 2100ABC. As a result, the calculated methane global lifetime is unchanged in the future and at present. The impact of climate change is to increase both the ozone photochemical production and destructions terms. Overall, the increased destruction dominates and the net chemical production is reduced by 308 Tg/yr from exp. 2100AB to 2100ABC. In the mid and upper troposphere, between 50 S and 50 N, the climate change is responsible for an ozone increase reaching 8%. This feature has two major origins. First, as already obtained by Zeng and Pyle [2003] but to a much lesser extent in LMDz-INCA, an enhanced meridional circulation is calculated in a future climate. As shown in Table 1, the stratospheric influx increases by 175 Tg/yr from exp. 2100AB to 2100ABC. Second, in a warmer climate, the hydrological cycle is more active and the convection is enhanced. Due to the more intense convective activity, the calculated lightning NO_x emission increases by 50%, from 5 Tg(N)/yr to 7.5 Tg(N). Consequently, the net ozone production increases in the tropics above about 400 hPa up to the tropopause. In the upper troposphere at high latitudes an ozone decrease of 6–10% is calculated as a result of variation in the tropopause height. At these

latitudes, the tropopause pressure decreases by 15–25 hPa due to climate change, in agreement with Collins *et al.* [2003].

[7] Figure 2 depicts the surface ozone distribution calculated for exp. 2100ABC along with the deviation from present-day distributions associated with the different processes. During summer in the northern hemisphere, surface ozone reaches values generally larger than 70 ppb over northern America, western Europe, northern Africa and the Middle East as well as northern India and China (Figure 2a). As shown in Figure 2b, in these regions the ozone change associated with all processes (exp. 2100ABC) shows a strong increase reaching more than 30 ppb. A maximum increase reaching 55 ppb is calculated over the Middle East. Ozone is significantly increased over the whole tropical region due to a dramatic increase in fossil fuel and biofuel emissions. In the southern hemisphere, the ozone increment is transported away from source regions leading to an increase of 10–15 ppb over the South Atlantic, the Indian and even Pacific oceans. Figure 2c shows that biogenic emissions contribute significantly to the ozone increase calculated in the northern hemisphere (exp. 2100AB–2100A). Over northern America, western Europe, and northern China, surface ozone increases by up to 15–20 ppb due to increased biogenic emissions. A strong increase is also calculated over the Middle East where ozone precursors emitted over Europe tend to accumulate. Over these regions, biogenic emissions contribute by 30–50% to the future ozone increase. The ozone change (exp. 2100ABC–2100AB) associated with climate change looks more heterogeneous (Figure 2d). Over the ocean, where the

ozone destruction due to water vapour prevails, ozone decreases by up to 8 ppb. Over the continents, ozone increases by up to 8–10 ppb over the eastern US, western Europe and China. In the tropics, ozone also increases by up to 10 ppb over South America and Africa. The change in surface ozone appears strongly correlated with the temperature increase over the continents. This feature is mainly a result of enhanced PAN thermal decomposition which causes this species to decrease by up to 50% over source regions and ozone net production to increase. A boundary layer tracer has been introduced in the model to assess the role played by dilution processes. The more vigorous convection predicted in a warmer climate has a moderate impact outside the tropics. We find however, but to a much lesser extent than Rind *et al.* [2001], an enhanced continental ventilation and increased lightning NO_x emission over the central US. In contrast, more stagnant conditions are calculated over western Europe and the eastern US increasing the level of pollutants in these regions. An enhanced monsoon over India and Southeast Asia increases the boundary layer venting over these regions and a more intense outflow of pollution to the Pacific ocean is calculated.

[8] We calculate a tropospheric ozone net radiative forcing (Table 1) from the present to 2100 of 0.52 Wm⁻² due to anthropogenic emissions (exp. 2100A). Accounting for the impact of biogenic emissions increases the forcing to a value of 0.58 Wm⁻² (exp. 2100AB). Interestingly, considering the climate change also causes the forcing to slightly increase even if the ozone burden is reduced by 6 Tg from exp. 2100AB to 2100ABC. As shown in Figure 2b, the result of climate change is indeed to increase ozone in the upper troposphere where the impact on the forcing is enhanced. Climate feedbacks and biogenic emissions increase the future ozone radiative forcing by 12%. This positive feedback contrast with the overall negative feedback found by Stevenson *et al.* [2000] and with the strong positive feedback calculated by Zeng and Pyle [2003], and is clearly in between these two previous studies.

5. Conclusion

[9] In this study we applied a state-of-the-art climate-chemistry model and a biogenic emission model to investigate the relative impacts of anthropogenic and biogenic emissions and global warming on the evolution of ozone in 2100. Increased emissions cause ozone to build up in the whole troposphere. Climate change partly offsets this increase below about 500 hPa as a result of enhanced ozone photochemical destruction in a warmer and wetter climate. In the upper troposphere, ozone is further increased between 50 S–50 N when climate change is considered. A more intense Brewer-Dobson circulation causes the stratospheric ozone influx to increase by 35% and a more vigorous convective activity induces an increase of lightning NO_x emissions from 5 to 7.5 Tg(N)/yr. As a result of these competing effects, climate change slightly decreases the global tropospheric ozone burden. Due to the counteracting effects of increased CO and CH₄ causing OH to decrease in the future and of NO_x and enhanced water vapor causing OH formation to increase, the global

and annual mean OH concentration remains quite stable over the 21st century and the global methane lifetime is unchanged in the future run. The build up of surface ozone is strongly correlated with the temperature increase. The continental warming affects the PAN thermal decomposition over source regions with a direct impact on the NO_x levels and ozone chemical production. In addition to that, the more stagnant conditions calculated over western Europe and the eastern US favor the development of more intense pollution episodes. Increased biogenic emissions play a crucial role in the future development of ozone pollution. Their contribution increases surface ozone by 30–50% in northern continental regions during summer. The global increase of tropospheric ozone during the course of the century induces an additional radiative forcing of 0.58 Wm⁻². The feedback of climate change and of biogenic emissions causes this forcing to be 12% higher than the one calculated with anthropogenic emissions solely.

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