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Role of climate feedback on methane and ozone studied with a coupled Ocean-Atmosphere-Chemistry model.

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Abstract. We present results from two experiments carried out with a coupled ocean-atmosphere-tropospheric chemistry model run continuously over the period 1990 to 2100. In the control experiment, climate is unforced, but emissions of trace gases to the chemical model increase in line with an illustrative scenario for future trace gas emissions with medium high growth. In the climate change experiment trace gas emissions are identical to the control, but climate is also forced using greenhouse gas concentrations and SO₂ emissions from the same scenario. Global average methane in the climate change experiment increased from 1670 ppbv in 1990 to 3230 ppbv by 2100, compared to 3650 ppbv by 2100 in the control. The methane increase in the control experiment is therefore 27 % more than in the control. This difference is due to both temperature and OH changes which increase the rate of methane oxidation and act in the opposite direction to the negative feedback of methane on itself through OH. Mid-latitude northern hemisphere ozone concentrations in July for the mid-troposphere rose from 39 ppbv in 1990s to 64 ppbv in the 2090s in the control experiment and to 49 ppbv in the climate change experiment. The direct role of climate change is therefore predicted to be a negative feedback on the radiative forcing from the change to tropospheric ozone and methane concentrations.

Introduction

The importance of climate change to tropospheric chemistry has been recognised for some time, for example *Thompson et al.* [1989] applied increases in water vapour concentrations derived from climate change experiments to test box model sensitivity. Later, two-dimensional model experiments by *Fuglestedt et al.* [1995] also found decreases to ozone and increases to OH in response to increases in temperature and humidity. More recently *Johnson et al.* [1999], *Grewe et al.* [1999], and *Stevenson et al.* [2000] have used three-dimensional models in time-slice experiments to show that climate changes reduce ozone concentrations and decrease the methane lifetime. The future course of tropospheric methane concentrations is therefore expected to depend on its lifetime, affected by both the physical and chemical environment, as well as on future emissions. Increasing concentrations of carbon monoxide and of methane itself are expected to decrease OH and therefore increase the lifetime of methane [*Prather, 1994*]. Increases to NO_x and ozone concentrations will have the opposite

effect in that they react with HO₂ to produce increases to OH mainly over continental areas [*Johnson et al., 1999*]. Water vapour increases will affect methane through increases in the production of OH from the reaction of O(¹D) with water vapour, and increases to temperature will increase the rate of reaction of methane with OH. Here we present the results from two 110 year simulations of a global 3-D atmosphere-ocean-chemistry model to demonstrate the feedbacks between climate change and tropospheric ozone and methane concentrations.

Coupled Ocean-Atmosphere-Tropospheric Chemistry model

The Hadley Centre atmosphere-ocean GCM (AOGCM) was coupled to the tropospheric chemistry model STOCHEM every three hours. At each coupling step, the fields of wind, temperature, specific humidity, cloud amounts and positions, precipitation, boundary layer depth, surface heat flux and pressure are passed to STOCHEM. The AOGCM version used was HadCM3 ([*Gordon et al., 2000*]) which has a horizontal resolution of 2.5° × 3.75°, with nineteen layers in the vertical. This model does not use flux adjustment. STOCHEM is the Met. Office Lagrangian chemistry-transport model, which uses horizontal resolution of 5.0° × 5.0°, with nine layers between the surface and 100 hPa. STOCHEM is described in detail by *Collins et al.* [1997, 1999]; these and other studies [*Kanakidou et al., 1999a, 1999b*] show validations against observations of O₃, NO_x, CO, and HO_x pre-cursors. The model version used here differed from *Collins et al.* [1999] in the following ways. The rate coefficient for the reaction between OH and methane was taken from *Dunlop and Tully* [1993]. This rate coefficient is around 3 % less than more recent evaluations [*DeMore et al., 1997*], but was retained here to ensure compatibility with previous results. The chemical scheme was reduced to fifty species by removing all the sulphur species and the aromatic hydrocarbon chemistry. Other details of the model are as described by *Stevenson et al.* [2000].

Experiments

Two experiments were developed: a climate change and a control experiment. The control experiment used the control HadCM3 AOGCM, with constant greenhouse gases set at their pre-industrial values but in the climate change experiment greenhouse gases evolved in accordance with the A2 scenario from the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES) [*Nakićenović et al., 2000*]. Trace gas emissions used by STOCHEM in both experiments were as prescribed by the SRES A2 scenario, i.e. the atmospheric composition

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of HadCM3 and STOCHEM were uncoupled except for the methane concentration in the climate change run which was determined in STOCHEM for use in HadCM3. The SRES A2 scenario determines the spatio-temporal evolution of anthropogenic emissions of NO_x , CO, CH_4 and non-methane hydrocarbons (NMHC) in STOCHEM. Natural emissions, including the NO_x source from lightning were kept constant in both experiments. Other details of the emission scenario were essentially the same as described in *Stevenson et al.* [2000].

Results and Discussion

Figure 1a shows that the globally and annually-averaged temperature at 1.5m above the surface increased by 3.5°C between the decade 1990-9 and 2090-9 for the climate change experiment. The temperature increase was larger in the upper troposphere, at around 5.0°C . Table 1 shows the associated rise in specific humidity for January and July as a zonal mean at various heights in the troposphere, and for three latitude ranges. The increase in specific humidity varies from 15 to 84 %, with the largest percentage increases in the upper troposphere.

Figure 1b shows changes to the globally-averaged methane concentration predicted for the years 1990 to 2100. In the control experiment the concentration increased from 1670 ppb in 1990 to 3650 ppb in 2099, but only to 3230 ppb in the climate change experiment. Most of the difference between the experiments occurs after 2040.

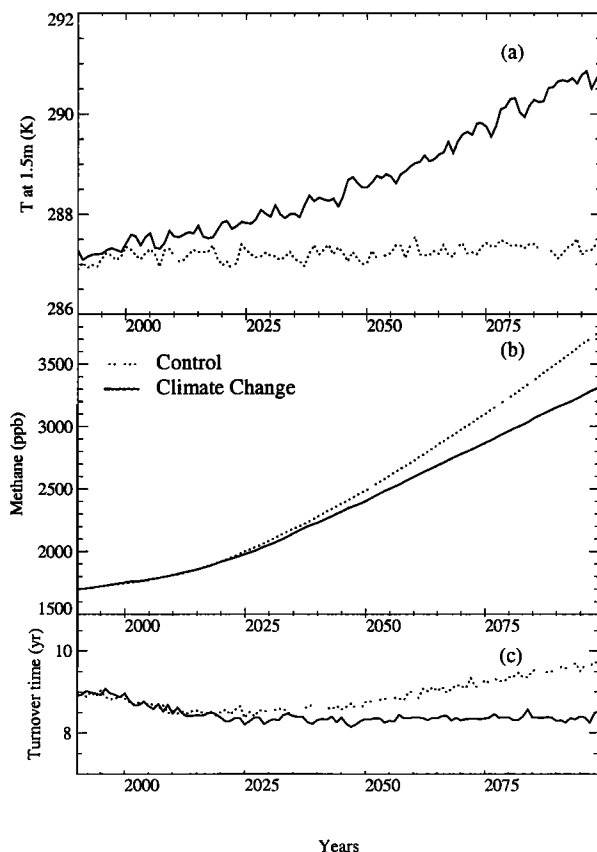


Figure 1. Global mean temperature at 1.5 m simulated over the period 1990-2100 in the control and climate change experiments (a), global mean methane concentrations (b), and the global mean methane lifetime in years (c).

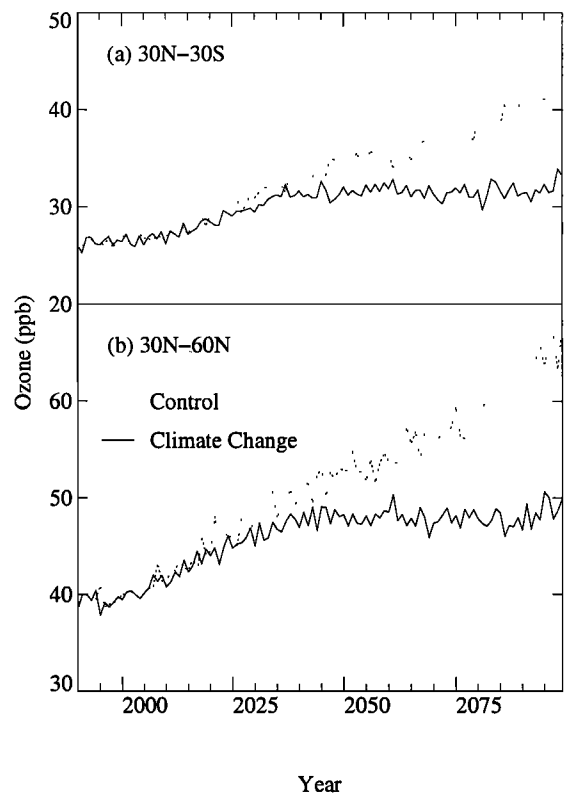


Figure 2. Zonal mean 650 hPa ozone concentrations for (a) 30°S - 30°N and (b) 30°N - 60°N in July simulated over the period 1990-2099 using control and SRES A2 climate experiments.

The lifetime for methane in both experiments is shown in figure 1c. For the control experiment the lifetime declines from around nine years in 1990 to around 8.5 years by 2025 and then increases to around 9.7 years by 2099. For the climate change experiment, the lifetime declines to around 8.3 years by 2025, but does not change significantly thereafter. Changes to the methane lifetime are produced from temperature change operating directly through the temperature sensitive rate coefficient for the reaction of methane with OH and through changes to OH itself. Both mechanisms are important as for example, the near-surface temperature rise for the 30°N - 30°S region implies a 7 % increase in the rate coefficient where OH changes between the two experiments are small.

Table 1 shows the zonal and decadal mean concentrations and changes to OH and water vapour between the decades 1990-9 and 2090-9 for three heights and three selected latitude ranges of the model. Hydroxyl radical concentrations in the control model generally decrease in all seasons and locations between the 1990s and 2090s due to the effect of emissions alone, with decreases generally between 2 and 10 %, though a 22 % decrease is found in January over the northern hemisphere. There is one exception, as OH increased at 350 hPa for July in the southern hemisphere. With the climate change experiment, the combined effect of emission increases together with climate change produces changes in OH concentrations which range from 12 % decreases to 12 % increases.

The influence of the feedback from climate change is complex. In most seasons and heights the effect of climate

Table 1. Zonal and decadal-averaged OH and Specific Humidity (Q) for control and climate change experiments for 1990-9 (volumetric mixing ratio $\times 10^{14}$ for OH and in $\text{g/kg} \times 10^3$ for specific humidity), and the percentage change to 2090-9 (in parentheses) for three latitude ranges.

	Control						Climate Change					
	30N-60N		30N-30S		30S-60S		30N-60N		30N-30S		30S-60S	
<i>January</i>												
OH, 950 hPa	1.49	(-23)	7.55	(-8)	3.17	(-2)	1.48	(-12)	7.55	(-6)	3.13	(-6)
Q, 990 hPa	1.90	(4)	13.4	(2)	5.02	(2)	1.90	(40)	13.6	(19)	5.12	(16)
OH, 650 hPa	1.90	(-12)	12.3	(-11)	7.77	(-10)	1.89	(2)	12.6	(-4)	7.72	(-1)
Q, 700 hPa	0.82	(2)	4.39	(3)	1.67	(2)	0.83	(37)	4.50	(24)	1.67	(29)
OH, 350 hPa	2.46	(-6)	10.6	(-9)	8.04	(-6)	2.44	(9)	10.7	(10)	8.18	(1)
Q, 350 hPa	0.05	(6)	0.42	(6)	0.16	(6)	0.06	(62)	0.45	(58)	0.16	(54)
<i>July</i>												
OH, 950 hPa	9.81	(-5)	7.78	(0)	1.02	(-9)	9.57	(10)	7.87	(2)	1.07	(-9)
Q, 990 hPa	6.85	(2)	14.1	(1)	3.21	(2)	6.72	(26)	14.3	(18)	3.32	(15)
OH, 650 hPa	17.5	(-12)	12.6	(-4)	1.35	(-3)	17.3	(-6)	12.7	(-4)	1.37	(-2)
Q, 700 hPa	3.03	(1)	4.90	(2)	1.01	(3)	2.97	(44)	4.99	(25)	1.05	(19)
OH, 350 hPa	16.1	(-11)	11.3	(-4)	1.59	(5)	15.9	(8)	11.4	(12)	1.59	(10)
Q, 350 hPa	0.27	(3)	0.45	(4)	0.06	(6)	0.26	(84)	0.46	(67)	0.06	(46)

change is to increase OH concentrations, though the opposite occurs in the southern hemisphere summer for near-surface concentrations. The largest increases from climate change of up to 19 % are at 350 mb, where the percentage changes to water vapour are also greatest. The northern hemisphere has consistently larger percentage increases to OH from climate change for the near surface and mid troposphere, with an increase of 15 % in July near-surface OH. In contrast, near surface tropical OH concentrations are not significantly affected by climate change.

Figure 2 shows the simulated ozone concentrations in July at 650 hPa for the years 1990 to 2100 for the tropical and subtropical region (30 °S-30 °N) and for the northern hemisphere midlatitudes (30 °N-60 °N). For the control experiment the increase in the decadal average of ozone between the 1990s and the 2090s was 15 ppb for the tropics and 26 ppb for the northern hemisphere midlatitudes, whereas it was only 6 ppb and 9 ppb respectively for the climate change experiment. Significant decreases to the zonally averaged ozone concentration due to climate change feedbacks are apparent in almost all seasons and heights, and globally averaged ozone changes between the 1990s and the 2090s were 16.2 ppbv in the control experiment and 8.5 ppbv in the climate change experiment. Although there is less methane in the climate change model, this is not the cause of lower ozone concentrations in this simulation as there is almost no difference in the flux of $\text{CH}_3\text{O}_2 + \text{NO}$ between the two models in the 2090s, and the total chemical production is slightly reduced by 20 Tg/yr in the climate change experiment. However the loss of ozone through $\text{O}(^1\text{D}) + \text{H}_2\text{O}$ is 26 % larger in the climate change experiment, and the net chemical production in the model reduces from 1060 Tg/yr to 880 Tg/yr for the period 2090-9. In the same period, the input of ozone at the top of the model domain increases from 360 to 420 Tg/yr due to changes in circulation at this level in the climate change experiment, so there is a decrease of

around 120 Tg/yr in the net production of ozone due to the impact of climate change. These changes to the ozone input at the top of the model must be treated with caution as they reflect only the change in circulation in the GCM without considering feedbacks to Stratospheric ozone concentrations. The changes to the ozone budget are similar to those presented by [Stevenson *et al.*, 2000], for time-slice experiments, though in that case the ozone chemical production increased by 160 Tg/yr as a result of climate change. This difference is presumably due to the fact that the time-slice experiments used a constant initial methane concentration and could not therefore allow for climate feedback on methane.

Conclusions

Calculation of future methane concentrations without considering climate changes resulted in an overestimate of 420 ppbv in the concentration by 2100. This change was brought about by significant differences in the methane lifetime between the two experiments after 2025. Globally averaged tropospheric ozone in the 2090s was decreased by around 8 ppbv by the effect of climate change mainly through reduction of the net production of ozone in the model. The negative climate change feedback shown here for the latter part of this century indicates that current estimates of the buildup of methane and tropospheric ozone to 2100 may be greatly overestimated. The role of climate change on natural emissions, including those of methane itself was not considered here, and for example, the NO_x source from lightning represents a positive feedback between climate change and ozone [Toumi *et al.*, 1996] that could affect these conclusions. Another source of uncertainty is in the parameterisation of the ozone input from the stratosphere at the top of the model domain. This does not include any feedback between dynamics and ozone concentrations, and further work is needed to improve understanding

of stratosphere-troposphere exchange which is highly variable between different model representations and is an important term in the ozone budget of the upper troposphere [Collins *et al.*, 2000].

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