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Future estimates of tropospheric ozone radiative forcing and methane turnover - the impact of climate change

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Abstract. We present a range of estimates for future radiative forcings due to changes in tropospheric ozone (O_{3T}). Ozone distributions were generated by the UKMO 3-D chemistry-transport model for 1990, 2030, 2060, and 2100, using four sets of boundary conditions. Anthropogenic emissions evolved following either the IPCC SRES "high" (A2) or "central" (B2) case. Each scenario was run with both a fixed (1990) climate, and with a changing climate, as generated by a coupled ocean-atmosphere GCM, forced with IS92a emissions. Calculated global mean O_{3T} radiative forcings for the A2 (B2) cases for 1990-2100 were +0.43 (+0.22) W m^{-2} when climate change was ignored; these fell to +0.27 (+0.09) W m^{-2} when climate change was included. Without climate change, CH_4 lifetimes (τ_{CH_4}) lengthened by 7-12 % between 1990 and 2100; however, when climate change was included, τ_{CH_4} fell by 0-5 %. Hence climate warming exerts a negative feedback on itself by enhancing O_{3T} and CH_4 destruction.

Introduction

Since pre-industrial times, increases in tropospheric ozone (O_{3T}) are estimated to have caused a radiative forcing on climate of +0.35 ± 0.15 W m^{-2} [*WMO*, 1999], whilst increases in CH_4 have contributed about +0.5 W m^{-2}; these compare to a CO_2 forcing of +1.6 W m^{-2} for the same time period [*Houghton et al.*, 1996]. Future O_{3T} and CH_4 concentrations will depend upon both emissions [*van Dorland et al.*, 1997] and climate change [*Johnson et al.*, 1999], since tropospheric chemistry is influenced by water vapour, temperature, cloud amounts, and atmospheric dynamics. Natural emissions, such as lightning NO_x and hydrocarbons from vegetation, are also likely to increase in a warmer, more CO_2-rich atmosphere, tending to increase O_{3T}. Here, we have fixed natural emissions, and studied the effects of anthropogenic emissions increases and climate change on O_{3T}, its radiative forcing, and the CH_4 lifetime (τ_{CH_4}).

Chemistry-transport model

We used the UK Met. Office (UKMO) chemistry-transport model STOCHEM, driven by meteorology from the Hadley Centre GCM. STOCHEM simulates 70 chemical species, at a horizontal resolution of 5°x5°, with nine equally spaced vertical levels 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 O_3, NO_x, CO, and HO_x pre-cursors. The model version used here differed slightly from *Collins et al.* [1999] in the following ways. Prescribed stratospheric ozone (O_{3S}) fields evolved using observed trends up to 1997 [*Forster*, 1999] and model predictions between 1997 and 2015 [*Austin et al.*, 2000]. The 2015 field was used for later years. The predicted partial O_{3S} recovery by 2015 in these runs means that there is only a small decline in O_{3S} between 1990 and future years. We also revised emissions (see below), updated quantum yields for O_3 photolysis to O^1D [*Michelsen et al.*, 1994], and used a new Eulerian dry deposition scheme.

Emissions

Two future trace gas emissions scenarios (A2 and B2) from the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES) (http://sres.ciesin.org) were used (Table 1). These scenarios describe the spatio-temporal evolution of anthropogenic emissions of NO_x, CO, CH_4 and non-methane hydrocarbons (NMHC). Scenario A2 has high population growth, moderate, regionally heterogeneous income and technology developments, and generates large emissions increases. Scenario B2 represents "dynamics as usual" [*Riahi and Roehrl*, 2000], with lower growth of emissions than A2. Both scenarios show strongest growth of emissions from the third world, with modest growth or reductions in emissions from the developed world.

The SRES scenarios lump biomass burning with other anthropogenic emissions, and give only annual distributions. Biomass burning emissions were extracted from the SRES totals, using ratios of biomass burning to other anthropogenic emissions from the EDGAR V2.0 database [*Olivier et al.*, 1996] for each 5°x5°grid square. Biomass burning emissions were then redistributed using monthly fields from *Cooke and Wilson* [1996]. Aircraft NO_x emissions were added, using NASA inventories for 1992, 2015 and 2050 [*Penner et al.*, 1999]. Natural emissions were as specified in *Collins et al.* [1999], except acetone, propane, and CO from vegetation, which were 20, 3.5, and 100 Tg yr^{-1} respectively, and oceanic CO (40 Tg yr^{-1}).

Future climate

Meteorological archives for the years 1990, 2030, 2060, and 2100, each with a 6-hourly resolution, were generated from a transient experiment of the Hadley Centre coupled ocean-atmosphere GCM (HadCM3) [*Johns et al.*, 1997], driven by greenhouse gas concentrations and other forcing

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Table 1. Total anthropogenic emissions and initial CH_4 concentrations for the A2(B2) scenario.

	NO_x $Tg(N)$ yr^{-1}	CO Tg yr^{-1}	$NMHC^a$ Tg yr^{-1}	CH_4^b ppmv
1990	29.9	893	138	1700
2030	60.8(48.4)	1410(1180)	210(202)	2179(2041)
2060	75.6(55.4)	1700(1470)	246(214)	2794(2395)
2100	110(60.5)	2480(2070)	295(170)	3943(2521)

^{a}NMHCs were split into individual species based on ratios taken from EDGAR [*Olivier et al.*, 1996].

^{b}Initial CH_4 concentrations were calculated by a 2-D model [*Johnson and Derwent*, 1996] driven by the appropriate emissions scenario.

factors derived from the IS92a emissions scenario [*Houghton et al.*, 1992]. The GCM run included all of the direct greenhouse gases, sulphate aerosol (direct and indirect effect), and O_{3T} from earlier runs of STOCHEM [*Stevenson et al.*, 1998]. Forced by IS92a, HadCM3 predicts global mean surface air temperature increases of 0.6, 1.5, and 3.1 K for the three future years relative to 1990. Tropospheric temperatures and humidities also increase, particularly in the tropics, and the Hadley Circulation intensifies. Future climate under SRES A2 and B2 scenarios will differ slightly from that predicted

under IS92a; analyses of HadCM3 runs [*Johns et al.*, 2000] indicate surface temperatures in 2100 are 0.5 K warmer (A2) and 0.8 K cooler (B2) than those predicted by IS92a.

Results and Discussion

A base case STOCHEM simulation was run using 1990 emissions and meteorology. For each of the years 2030, 2060, and 2100, four cases were run: using emissions scenarios A2 and B2 with 1990 meteorology, then with meteorology from the appropriate year of the IS92a climate. Each run was 15 months long, with the last 12 months analysed. Figure 1 shows changes in zonal annual mean O_{3T} since 1990 for each case. Scenario A2, with higher emissions than B2, produced larger increases in O_{3T}. Simulations including climate change showed smaller increases compared to those with a 1990 climate. Peak O_{3T} changes occur in the Northern Hemisphere upper troposphere (UT). The site of peak O_3 increases is controlled by the latitude of emissions increases (20-40°N) and the O_3 lifetime (τ_{O_3}), which is longest in the dry UT, particularly towards the poles. Ozone changes of most significance for radiative forcing are those in the UT over sub-tropical land, where there are few clouds, surface temperatures and albedoes are high, and the tropopause is high and cold. All cases show O_3 increases in these regions, although the B2 case with climate change shows tropical UT decreases by 2100, co-located with large increases in humidity, as also found by *Johnson et al.* [1999].
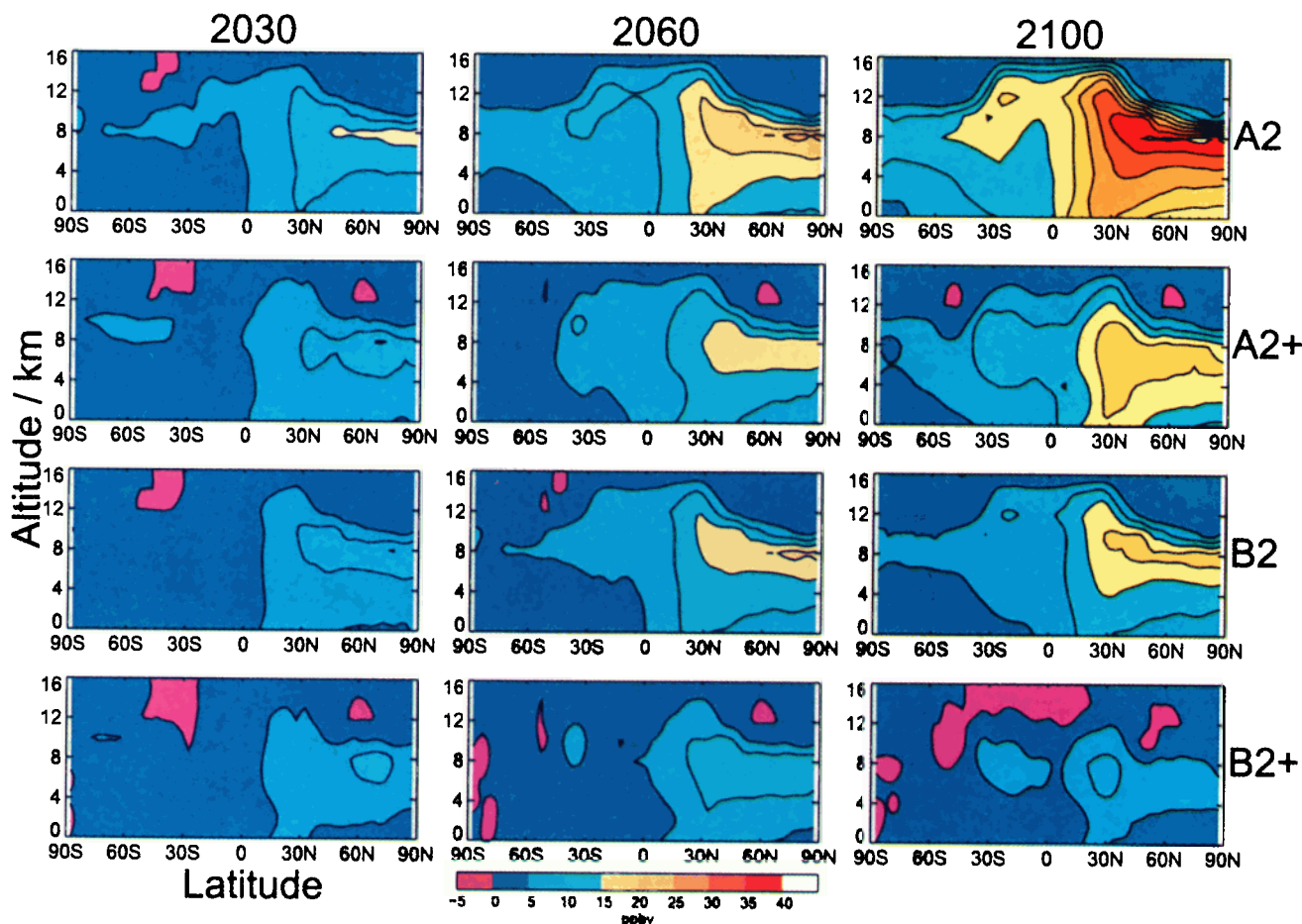
Figure 1. Zonal annual mean changes in O_{3T} since 1990 for the years 2030, 2060, and 2100, for the emission scenarios A2 and B2, using a 1990 meteorology and using a changing climate (+).
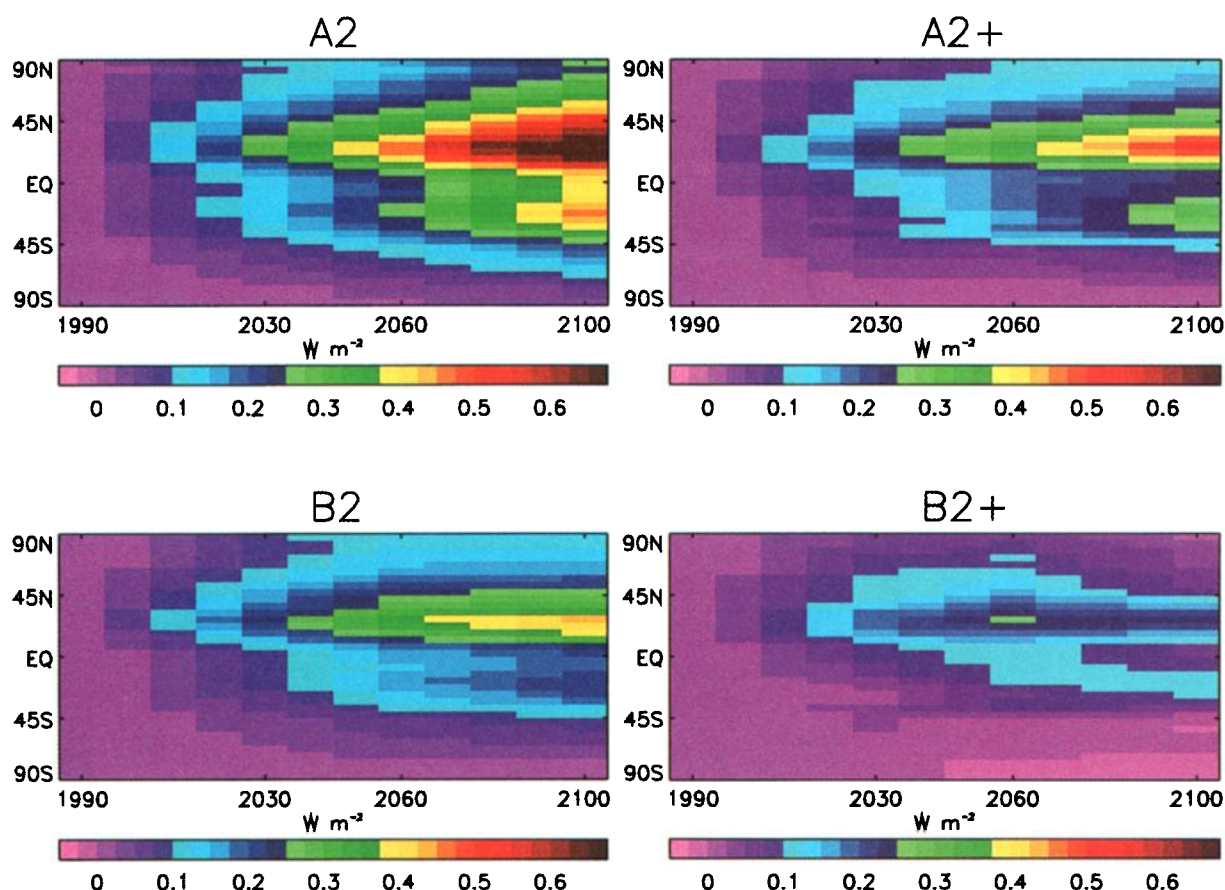
Figure 2. Evolution of zonal mean radiative forcing due to changes in O_{3T} since 1990, for the emission scenarios A2 and B2, using a 1990 meteorology and using a changing climate (+).

The impact of climate change can be understood in more detail from the global O_{3T} budgets (Table 2). Climate change increases chemical production (CP) of O_{3T}, but increases chemical destruction (CD) by more, leading to substantial reductions in net CP. Over 50 % of CD is through the reaction
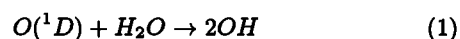
$$O(^1D) + H_2O \rightarrow 2OH \qquad (1)$$

Table 2. The 1990 global O_3 budget ($Tg(O_3)$ yr^{-1}) (TCP is total chemical production, TCD is total chemical destruction and NCP is net chemical production), O_3 and CH_4 lifetimes (τ), and percentage future changes, with (without) climate change, for emission scenarios A2 and B2. The final row is the global annual mean O_{3T} radiative forcing relative to 1990.

	1990	A2			B2		
		2030	2060	2100	2030	2060	2100
NO+HO$_2$	2841	39(38)	60(58)	104(100)	27(26)	41(39)	55(51)
NO+CH$_3$O$_2$	937	37(35)	67(61)	117(105)	31(28)	47(42)	50(40)
NO+RO$_2$	545	27(25)	37(35)	68(63)	21(20)	26(24)	20(14)
TCP	4323	37(36)	59(56)	102(96)	27(25)	41(38)	50(44)
Strat. Input	432	-6(-11)	2(-11)	2(-11)	-6(-11)	2(-11)	2(-11)
O(^{1}D)+H$_2$O	2305	32(27)	53(43)	91(70)	25(21)	40(30)	52(35)
O$_3$+HO$_2$	955	39(40)	67(69)	125(133)	28(28)	45(46)	55(59)
O$_3$+OH	381	27(28)	37(37)	47(51)	19(19)	25(25)	23(24)
TCD	3888	35(33)	57(52)	100(90)	26(23)	40(35)	51(41)
Dry Deposition	862	25(26)	36(41)	63(73)	17(18)	23(27)	22(30)
NCP	435	55(62)	71(93)	125(156)	39(45)	44(65)	43(69)
O$_3$ burden/Tg	316	13(16)	22(27)	32(49)	8(11)	14(19)	11(22)
τ_{O_3}/days	24.3	-16(-12)	-21(-15)	-32(-21)	-13(-9)	-17(-12)	-24(-12)
τ_{CH_4}/yrs	8.32	-6.7(-3.5)	-5.2(1.6)	0.0(12)	-5.3(-1.9)	-4.8(2.2)	-5.4(6.5)
$F_{O_3(T)}$/W m^{-2}	—	0.12(0.15)	0.19(0.26)	0.27(0.43)	0.08(0.11)	0.12(0.18)	0.09(0.22)

Climate warming increases H_2O concentrations, and increases the flux through reaction (1) by over 12 % in the 2100 cases. This perturbation is the dominant change to the O_{3T} budget resulting from climate change.

Table 2 shows that turnover of O_{3T} increases in future, as both emissions increases and climate change tend to increase the total CP and CD fluxes of O_{3T}. Future τ_{O_3} reduce by up to 32 %, so although there are large increases in net CP, these are not fully reflected in model O_{3T} burdens. Changes in stratospheric input are relatively small, however this process is highly simplified in STOCHEM. Dry deposition is a major sink for boundary layer O_3, and this term increases as concentrations rise. We used a fixed deposition velocity over land, although this will be a function of vegetation type and cover. Future changes in stratosphere-troposphere exchange and vegetation are potentially important climate-O_3 interactions needing further study.

Radiative forcings were calculated for the modelled O_{3T} changes, following the method described by *Stevenson et al.* [1998]. Figure 2 shows the evolution of the zonal mean forcing for each case, and global mean values are given in Table 2. The forcing peaks in the sub-tropics, with a significant growth in the Southern Hemisphere (SH), particularly in the A2 scenario, reflecting emissions increases. The impact of climate change is strong enough in the B2 scenario to reduce the forcing between 2060 and 2100 despite a general increase in emissions, and produces a negative forcing relative to 1990 in the polar SH.

These results also have implications for future CH_4 and its radiative forcing. In most cases τ_{CH_4} increases when emissions increase and climate change is ignored (Table 2). This is the view taken by *Houghton et al.* [1996], and reflects increased levels of CH_4 and CO, which depress OH, reducing the CH_4 sink. However, climate warming increases the temperature-dependant CH_4 oxidation rate coefficient [*Johnson et al.*, 1999], and increases in water vapour and NO_x concentrations tend to increase OH. In most cases these factors exceed the effect of rising CO and CH_4, and τ_{CH_4} falls when climate change is included (Table 2). Lower CH_4 levels will also reduce future O_{3T}. Long integrations of coupled chemistry-climate models are needed to calculate future trends of CH_4 and O_{3T} in a fully internally consistent way.

Conclusions

Future O_{3T} depends upon future pre-cursor emissions and climate change. Increases in emissions enhance net O_3 production, but climate warming, and in particular higher absolute humidity, tends to reduce net production. Inclusion of climate change reduces the global mean O_{3T} forcing by about 0.05 W m^{-2} per degree K of surface warming, lowering the 2100 estimates by 40-60 %. Climate change also tends to reduce τ_{CH_4} by about 5 % per degree K of surface warming. In this way, climate change exerts a negative feedback on itself through O_{3T} and CH_4.

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