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**Responses in Tropospheric Chemistry  
to Changes in UV Fluxes, Temperatures  
and Water Vapour Densities**

by

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# Responses in tropospheric chemistry to changes in UV fluxes, temperatures and water vapour densities

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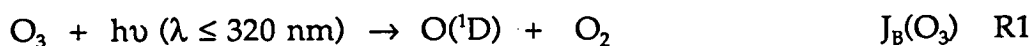
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**Abstract** A two-dimensional chemistry/transport model of the global troposphere is used to study the chemical response to i) increased UV radiation from stratospheric ozone depletion and ii) increased temperatures and water vapour densities that follow from increased levels of greenhouse gases. Increased UV radiation increases the photolysis rates for several tropospheric gases, in particular ozone. This leads to enhanced levels of odd hydrogen and reduced concentrations of tropospheric ozone. Increases in temperature and water vapour densities reduce the levels of tropospheric ozone through temperature dependent reaction rates and increased production of odd hydrogen. In both cases the methane levels are also reduced. Thus, the results indicate that the considered mechanisms constitute damping effects on global warming. For a global temperature increase of 3.88 K, the levels of tropospheric ozone and methane are reduced by about 10% and 18%, respectively.

## 1. Introduction

The oxidizing capacity of the troposphere is to a very large extent controlled by the levels of the extremely reactive hydroxyl radical, OH. Important trace gases such as CH<sub>4</sub> and other hydrocarbons, several halocarbons, NO<sub>2</sub> as well as sulphur containing species are removed by OH. In addition, OH also initiates the production of tropospheric ozone.

The initial production of OH occurs via the photolysis of ozone in the presence of water vapour:



Most of the excited oxygen atoms that are produced through R1 are energetically stabilized by air molecules (mainly N<sub>2</sub> and O<sub>2</sub>) to O(<sup>3</sup>P) which reacts with O<sub>2</sub> to yield ozone again. However, a few percent of the total loss of O(<sup>1</sup>D) occur through reaction

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with water vapour (R2) which produces two OH radicals. Since the rate of O(<sup>1</sup>D) production depends on the flux of UV, and stratospheric ozone controls the fluxes of UV radiation into the troposphere, the observed depletion of stratospheric ozone has increased the photodissociation of O<sub>3</sub> in the troposphere, and thereby the production of OH. This links tropospheric chemical changes to stratospheric ozone depletion, mainly caused by the CFCs (Liu and Trainer, 1988; Madronich and Granier, 1992; Fuglestvedt et al., 1994). There is also a potentially important link between greenhouse warming and tropospheric chemistry through temperature and water vapour increases. The initial effects will occur through temperature dependent reaction rate constants and through the water vapour dependent reaction R2.

The effects of stratospheric ozone depletion and climate change on tropospheric chemistry have been studied in earlier papers. Liu and Trainer (1988) used a box model with fixed concentrations of NO<sub>x</sub>, CO and hydrocarbons to study the response in O<sub>3</sub> and odd hydrogen to increased fluxes of UV. Thompson et al. (1989) used a one-dimensional model to study the effects of stratospheric ozone depletion and increased water vapour levels on O<sub>3</sub> and OH. De Leeuw and Van Rheineck Leyssius (1991) studied the sensitivity of episodic and long-term concentrations of O<sub>3</sub> in the boundary layer to changes in UV and temperatures using a Lagrangian long-range transport model. Based on observed reductions in total ozone, Madronich and Granier (1992) estimated the changes in the photolysis rate of O<sub>3</sub> that yields O(<sup>1</sup>D), J<sub>B</sub>(O<sub>3</sub>), focusing on the effects on OH and the growth rate of CH<sub>4</sub>. Madronich (1993) discusses the response in tropospheric chemistry to changes in UV. Fuglestvedt et al. (1994) used modelled changes in stratospheric ozone to study the responses in tropospheric chemistry, with focus on OH, CH<sub>4</sub> and O<sub>3</sub>.

In this paper we use a two-dimensional chemistry/transport model of the global troposphere to study how changes in UV, temperature and water vapour levels affect tropospheric chemistry, and focus will be given to O<sub>3</sub>, OH and CH<sub>4</sub>.

## 2. Model experiments

In the first set of model experiments, we study the time dependent responses to the increased UV radiation calculated from the observed reductions in total ozone from June 1979 through May 1994. In the second set, we focus on the steady state responses to modelled future changes in temperatures and water vapour. A two-dimensional (altitude vs. latitude) chemistry/transport model of the global troposphere is used to study the chemical responses to these changes. The model has 20 levels up to 16.25 km and a meridional resolution of 10°. Only gas phase chemistry is modelled, but first order heterogeneous scavenging of soluble species by droplets and sticking on aerosols are included. The chemistry is updated according to the latest compilations. See Fuglestvedt et al. (1994) for a further description of the model.

## 2.1 Effects of changes in stratospheric ozone on tropospheric chemistry

Total ozone levels from TOMS measurements are used. Starting with June 1979 until the end of 1992 we use the observations from the Nimbus-7 instrument. For the first 4 months of 1993 we use the average of the Nimbus-7 and the Meteor-3 instruments. For the remainder of 1993, and the first 5 months of 1994, measurements from the Meteor-3 instrument are used. Solar fluxes are calculated by an algorithm similar to the one described by Isaksen et al. (1977) with the modifications given by Jonson and Isaksen (1991). Photodissociation rates (J-values) for 16 photolytic reactions are calculated. Monthly (for the 15th of each month) J-values are calculated for every  $10^\circ$  latitude for each level in the chemistry/transport model with a diurnal variation using a time step of one hour. Climatological averages are applied for zonal cloud fractions, vertical distributions, cloud types and optical thickness (Lelieveld et al., 1989; Henderson-Sellers and McGuffie, 1987).

The satellite observations show a decrease in global annual total ozone of 8 % between 1979 and 1993. There are large seasonal and regional variations in the changes, and the reduction are largest at high latitudes during spring. (See Gleason et al. (1993) for an analysis of the trend from 1979 through 1992).

Figure 1 shows the observed development in global total ozone and the calculated development in model domain average of the dissociation rate of  $O_3$  that yields  $O(^1D)$  ( $J_B(O_3)$ ). Both are given as annual averages normalized to 1979 levels.

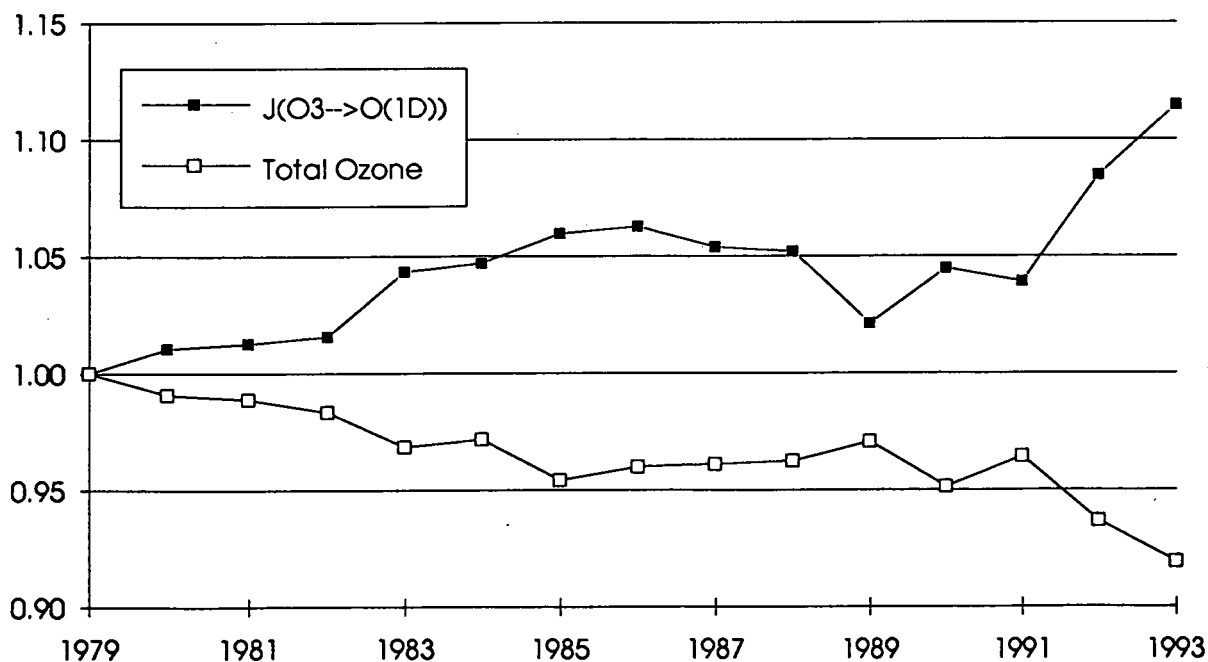


Figure 1. Development of observed global total ozone densities and calculated model domain average of  $J_B(O_3)$ , given as annual averages normalized to 1979 levels.

Figure 2 shows the calculated percentage increases in  $J_B(O_3)$  for 1993 and the first five months of 1994 relative to 1979 levels. The changes represent noon conditions and the lowest model layer (250 m) and are given as function of latitude and month. The figure shows large relative increases in  $J_B(O_3)$  at higher latitudes during spring, but also significant enhancements at lower latitudes. Due to the strong absorption by ozone in the wavelength region where  $O(^1D)$  is formed, the relative changes in  $J_B(O_3)$  are larger than the relative changes in total ozone.

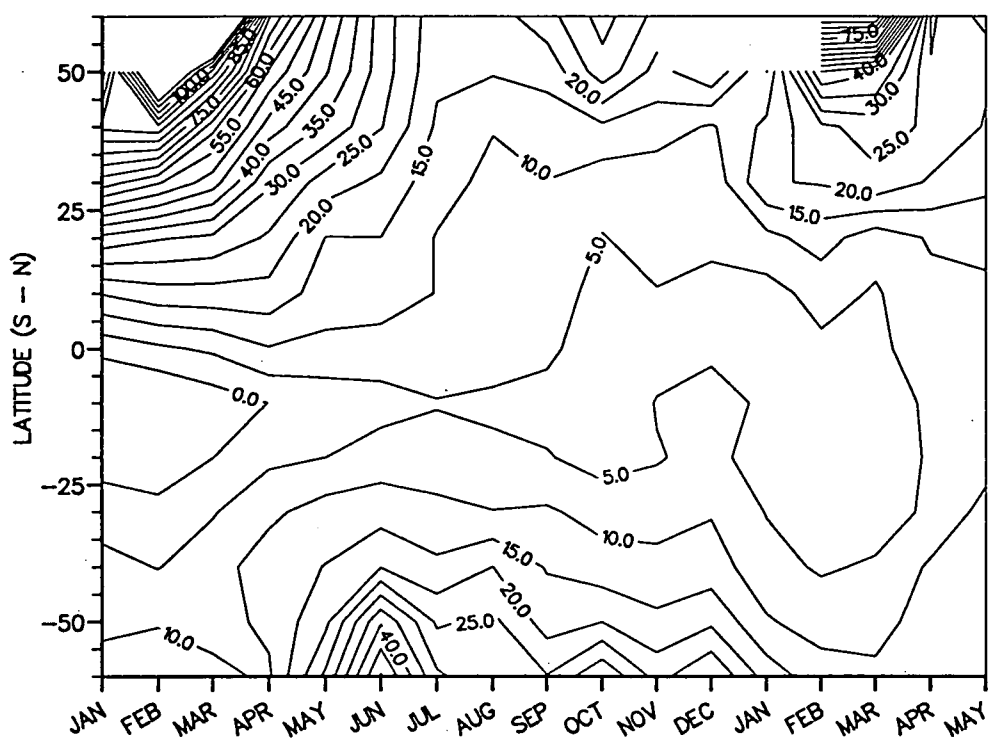


Figure 2. Calculated changes (%) in the noon values of  $J_B(O_3)$  in the lowest model layer for 1993 and the first five months of 1994 relative to 1979 levels.

The calculated changes in J-values for 16 photolytic reactions are applied to study the effects of the observed reductions in stratospheric ozone on the chemistry of the troposphere. (See Fuglestvedt et al. (1994) for a description of the photolytic reactions). Two model runs are performed: A reference run in which J-values representing the years 1979, 1980 and 1981 are used throughout the period June 1979 through May 1994. In addition, an increase of 0.7%/yr in the  $CH_4$  emissions is assumed. A perturbation run is then performed, in which the J-values are changed each month according to the changes in stratospheric ozone, with the same increase in the  $CH_4$  emissions as in the reference case.

The calculated response in OH to J-value changes shows largest percentage increase in the spring months when values are relatively low in the reference case. This means that in the perturbed atmosphere, high chemical activity starts earlier in the spring than in the unperturbed atmosphere. Figure 3 shows the calculated

response in OH to changed J-values for October as function of latitude and altitude. Large increases are found at high Southern latitudes. Figure 4 shows the annual cycles of OH for 50°N and 50°S (at 1750 m) in the reference and in the perturbed case. It is seen that the absolute increases at 50°N are larger than at 50°S, which is due to the more active chemistry at 50°N. The relative increases, however, are of similar magnitude.

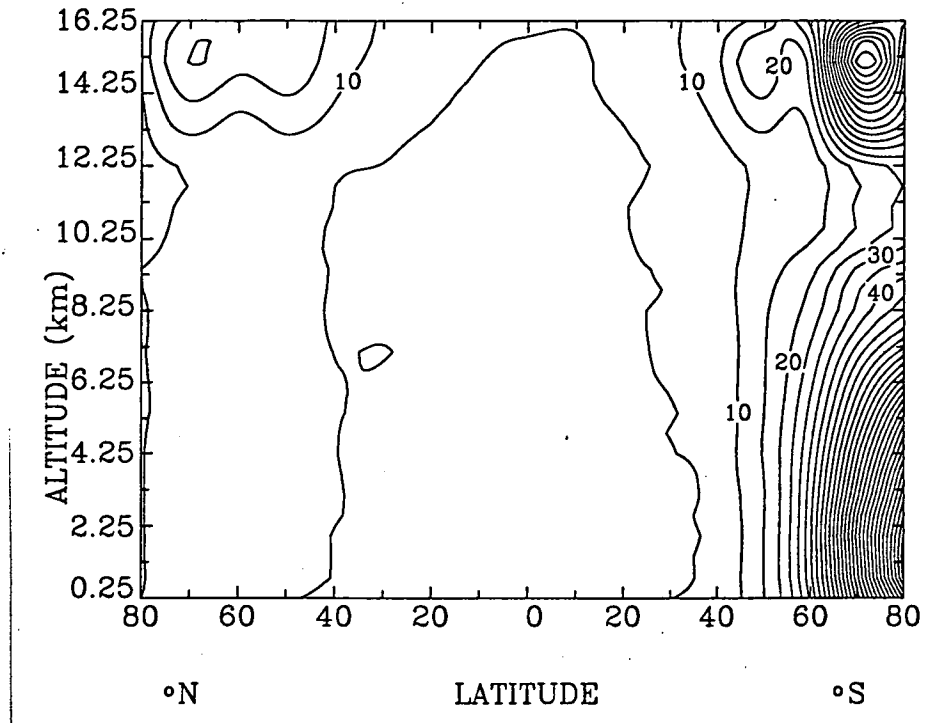


Figure 3. Calculated percentage increases in OH due to increased UV fluxes for October.

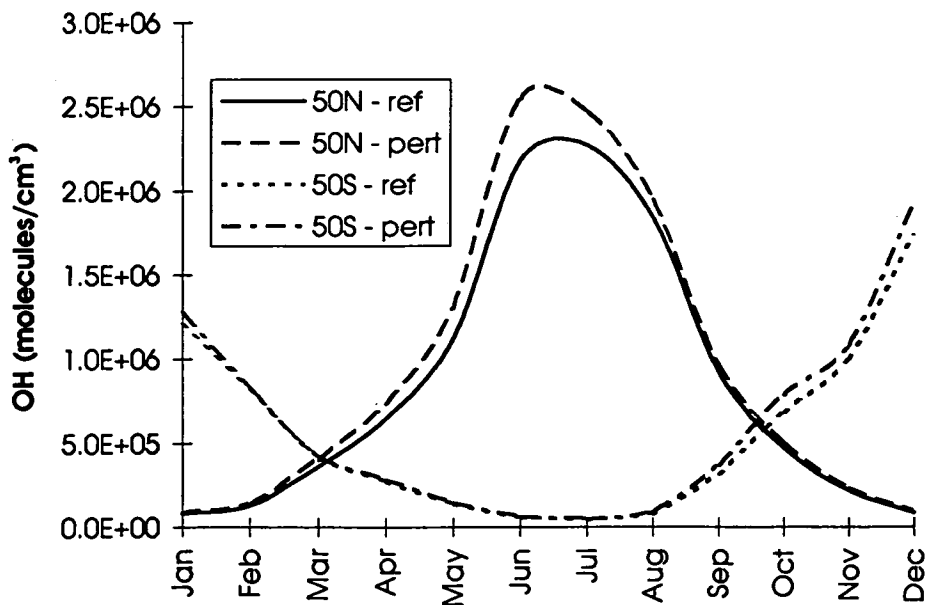


Figure 4. Calculated annual cycles in OH for the reference and the perturbed case.

Figure 5 and 6 show the calculated changes in tropospheric  $O_3$  between the two model runs as function of latitude and altitude for December and April, respectively. At high Southern latitudes there are large reductions in tropospheric  $O_3$  in response to the reductions in stratospheric ozone taking place in the austral spring. The maximum effect is delayed by about 2 months. At middle and high Northern latitudes in April, the increased UV radiation give increases in tropospheric  $O_3$  (figure 6), which is due to the high levels of  $NO_x$  and other ozone precursors (CO and hydrocarbons). This is the only month when we find extensive ozone increases. In May and June reductions in tropospheric  $O_3$  are calculated also for high Northern latitudes.

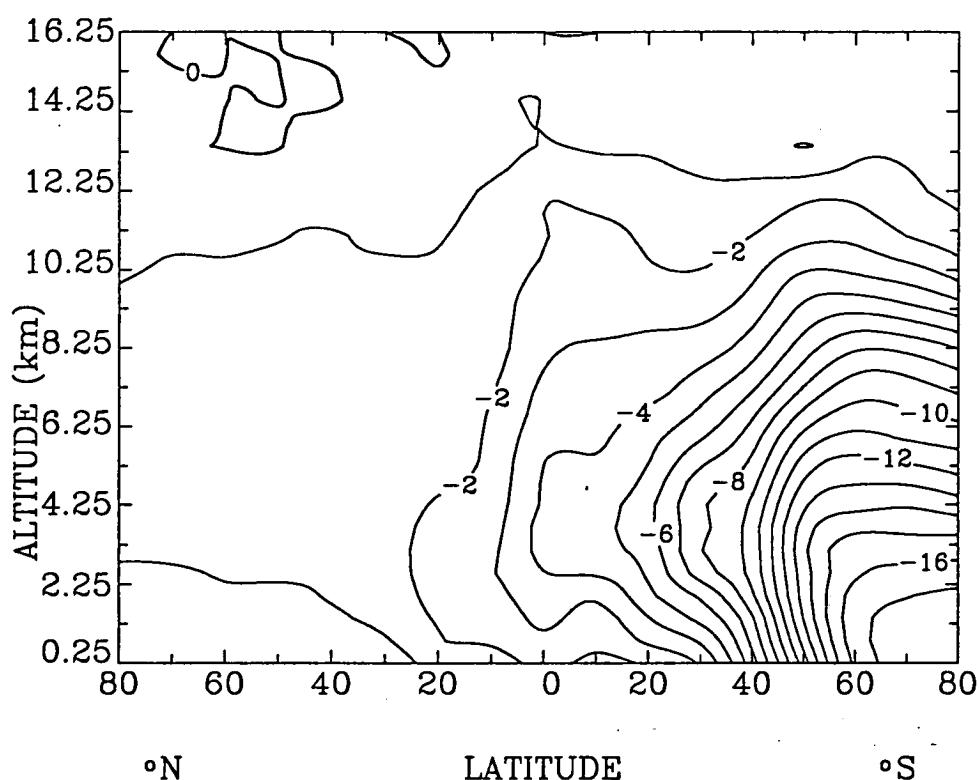


Figure 5. Calculated percentage changes in tropospheric  $O_3$  for December due to increased UV fluxes.

The calculations also show large year to year variations in the effects on tropospheric  $O_3$  reflecting the variations in ozone column densities. Figure 7 shows the percentage change in the annual levels of tropospheric  $O_3$  as a result of changes in UV. The numbers are given for the altitude level 4250 m and for four selected latitudes. The reductions in tropospheric ozone are largest at high Southern latitudes. The smallest changes are found at  $60^\circ N$ , and for 750 m at this latitude the trend is close to zero. The differences in the responses reflect the variations in the ozone production and loss processes.

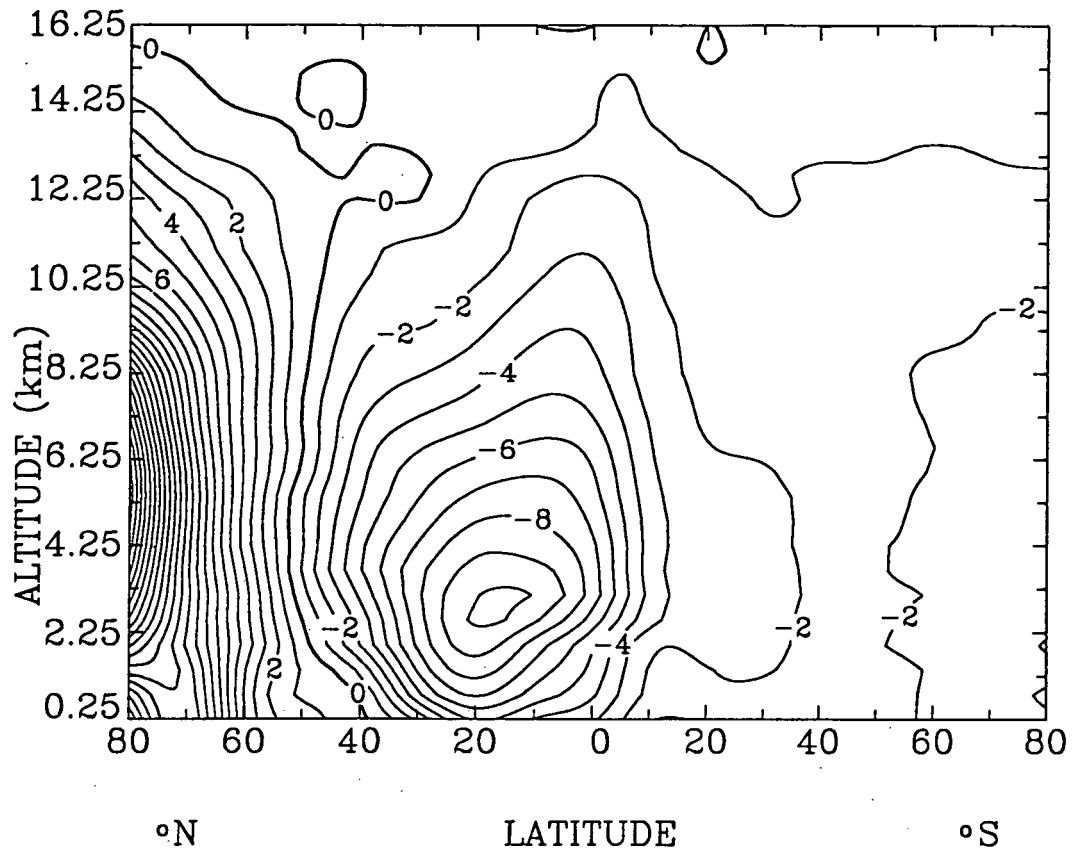


Figure 6. Calculated changes (%) in tropospheric O<sub>3</sub> for April due to increased UV fluxes.

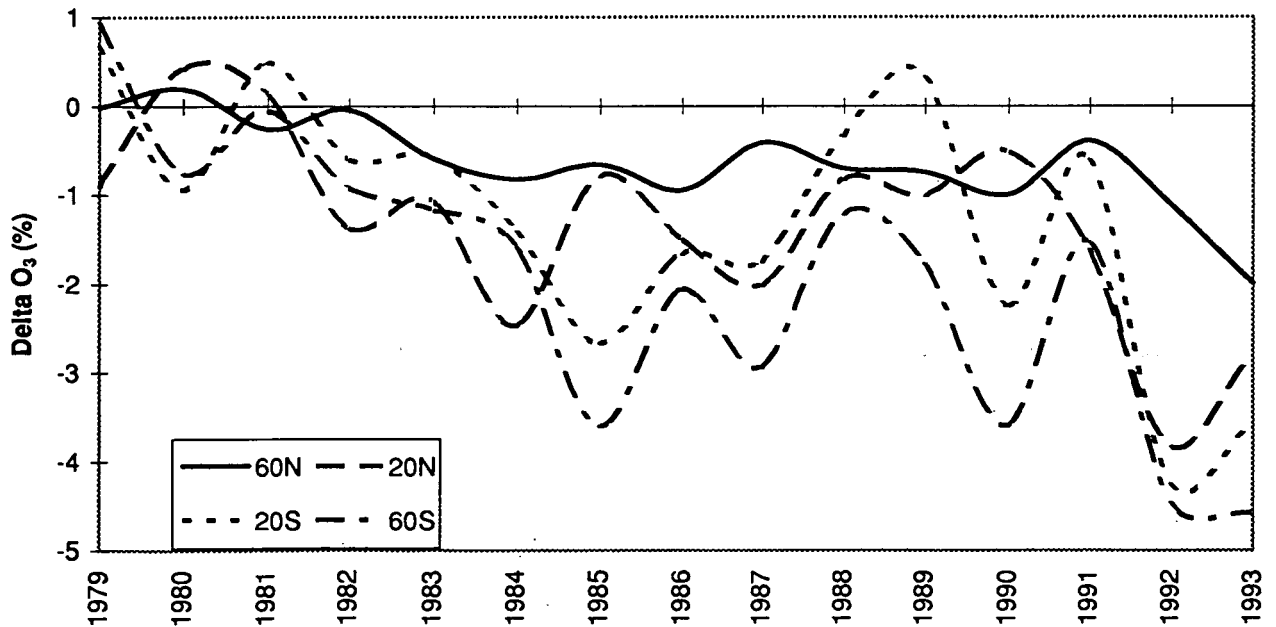


Figure 7. Accumulated reduction (%) in annual levels of tropospheric O<sub>3</sub> due to UV increases for 4250 m for selected latitudes.



Both CO and CH<sub>4</sub> have reaction with OH as their main sink. Since increased UV radiation gives higher levels of OH, increased loss is calculated for both gases. Figure 8 shows the change in global average level of CH<sub>4</sub> (in ppbv) as a result of changing UV radiation. The changes in the global annual growth rate (in ppbv/yr) are also plotted (right axis). The figure shows that the changes in UV reduce the global level by as much as 30 ppbv at the end of the period. In the mid 1980s the growth rate is reduced by 2-4 ppbv/yr, while during 1993/1994 the reduction reaches almost 7 ppbv/yr. For the latest years the calculations give somewhat higher reductions in the growth rate at high Northern latitudes than elsewhere.

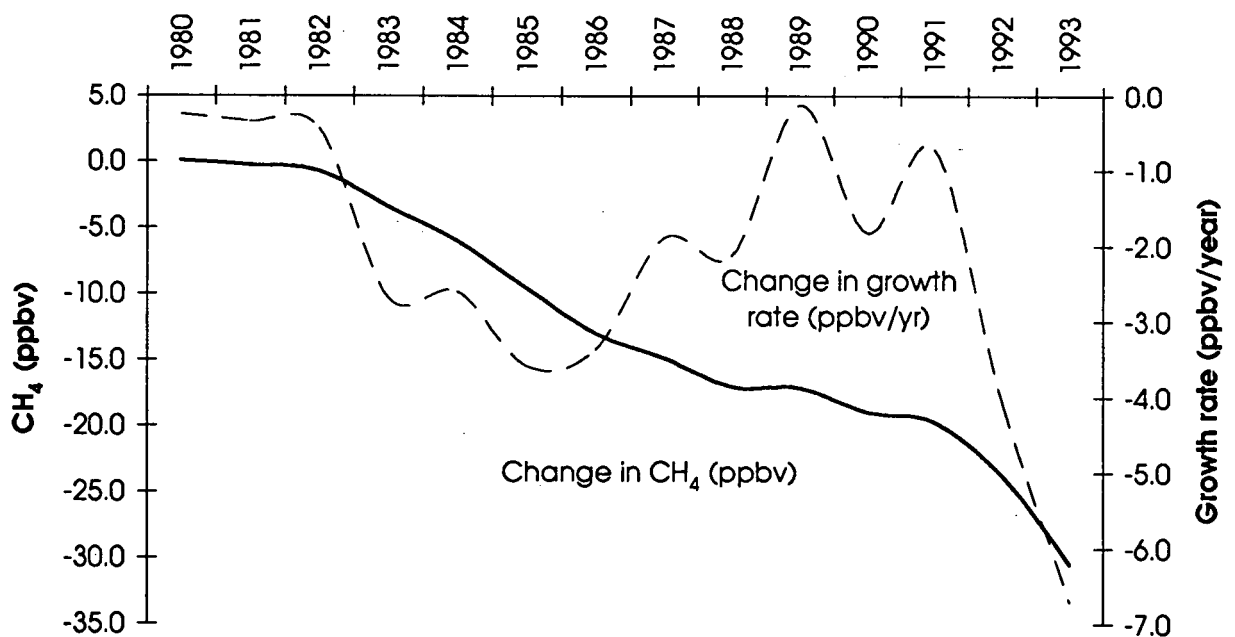


Figure 8. Changes in the global level and growth rate of CH<sub>4</sub> due to UV increases.

Figure 9 shows the percentage reduction in CO between the reference case and the case with changing UV. Moderate reductions are calculated, with largest effects at high latitudes.

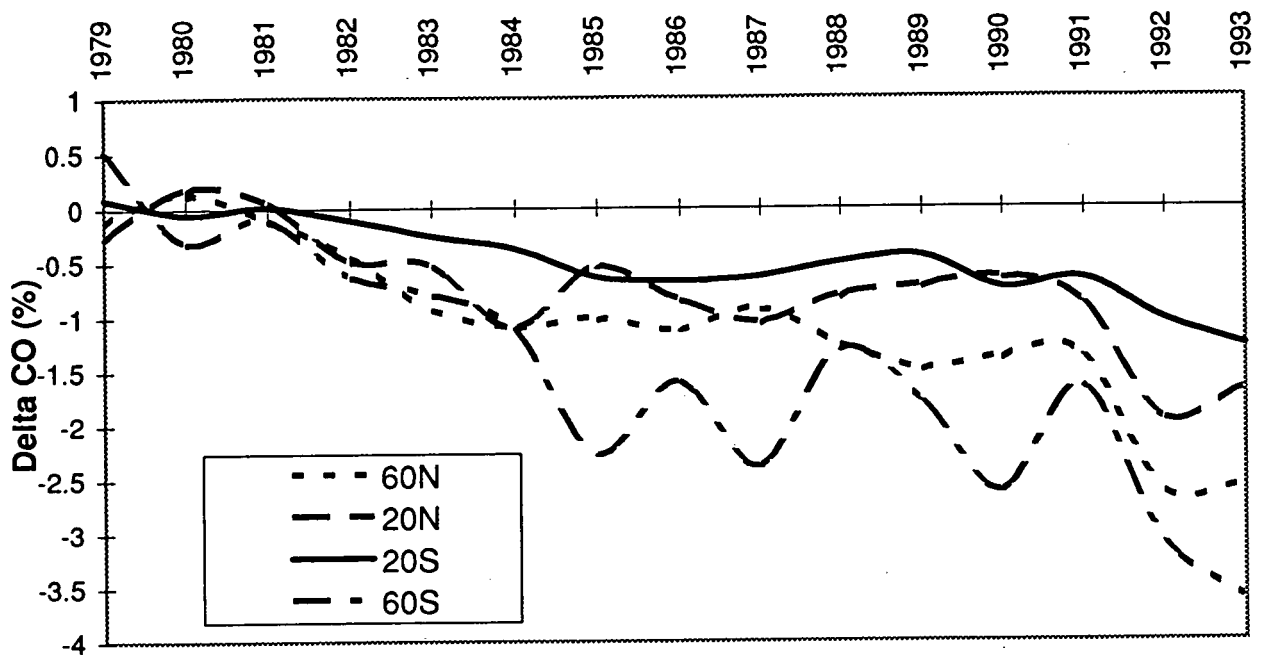


Figure 9. Accumulated reduction (%) in annual levels of CO at 750 m due to UV increases.

## 2.2 Effects of changes in tropospheric temperatures and water vapour densities

To study the effects of increased temperatures and water vapour levels, model results from a study by Wang et al. (1992) were applied. They used the 1990 observed levels of  $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ ,  $\text{CFCl}_3$  and  $\text{CF}_2\text{Cl}_2$  in modelling the reference climate. Then the individual gas concentrations were based on the Business-as-Usual scenario for 2050 from IPCC (1990). The modelled change in global annual mean radiative forcing of the troposphere-surface system is  $3.1 \text{ W/m}^2$ , while the increases in global annual mean surface temperature and column water vapour are 3.88 K and 27%, respectively. (No indirect effects from the CFCs through ozone changes were taken into account). The experiments were performed with a 3D climate model consisting of the National Center for Atmospheric Research community climate model version one for the atmosphere and a simple mixed-layer with sea ice for the oceans (see Wang et al., 1991). Figure 10 shows the calculated changes (K) in tropospheric temperatures between 2050 and 1990 as function of latitude and altitude for the winter season. The figure shows that the typical temperature increase is 3-4 K below  $\sim 5 \text{ km}$  south of  $50^\circ\text{N}$ . Larger increases are found over Equator between 7 and 13 km (5-6 K) and at high Northern latitudes (up to 12 K in the lowest level). The calculated percentage changes in water vapour for January are given in figure 11. Below 5-8 km and between  $50^\circ\text{N}$  and  $80^\circ\text{S}$ , the changes are generally in the range 20-40%.

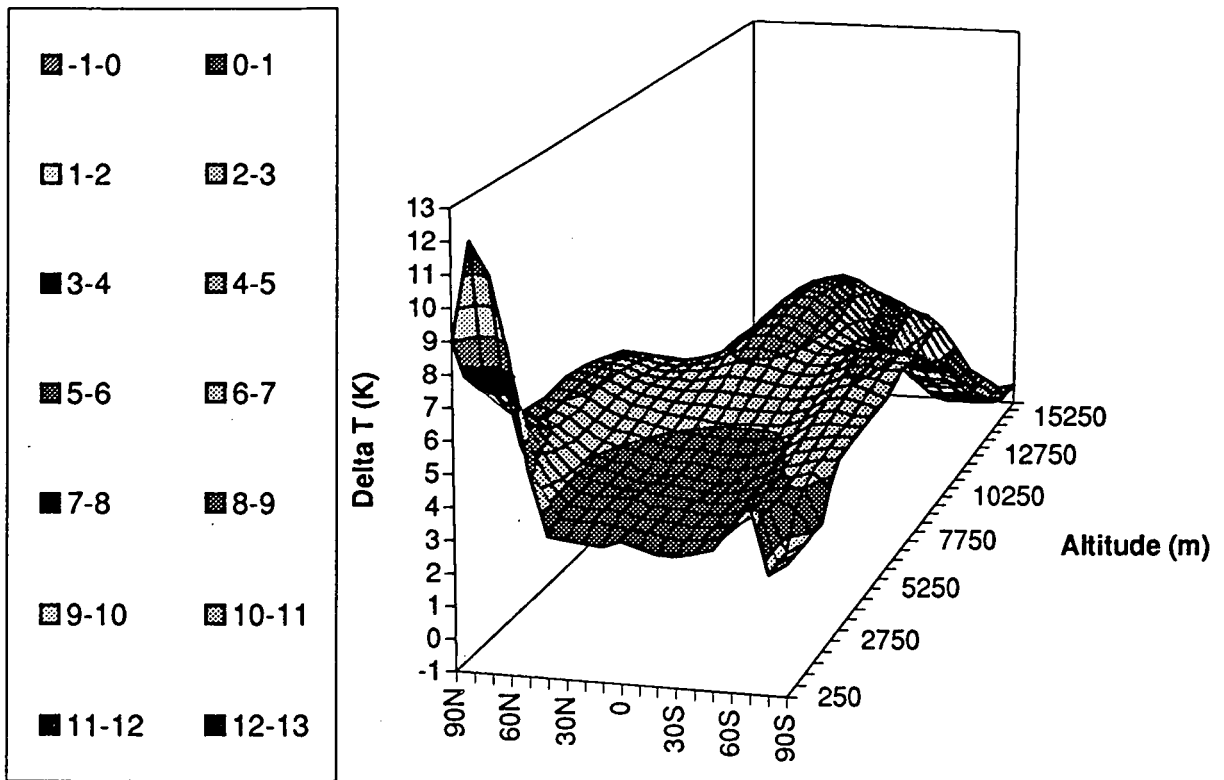


Figure 10. Calculated changes in tropospheric temperatures (K) between 2050 and 1990 as function of latitude and altitude.

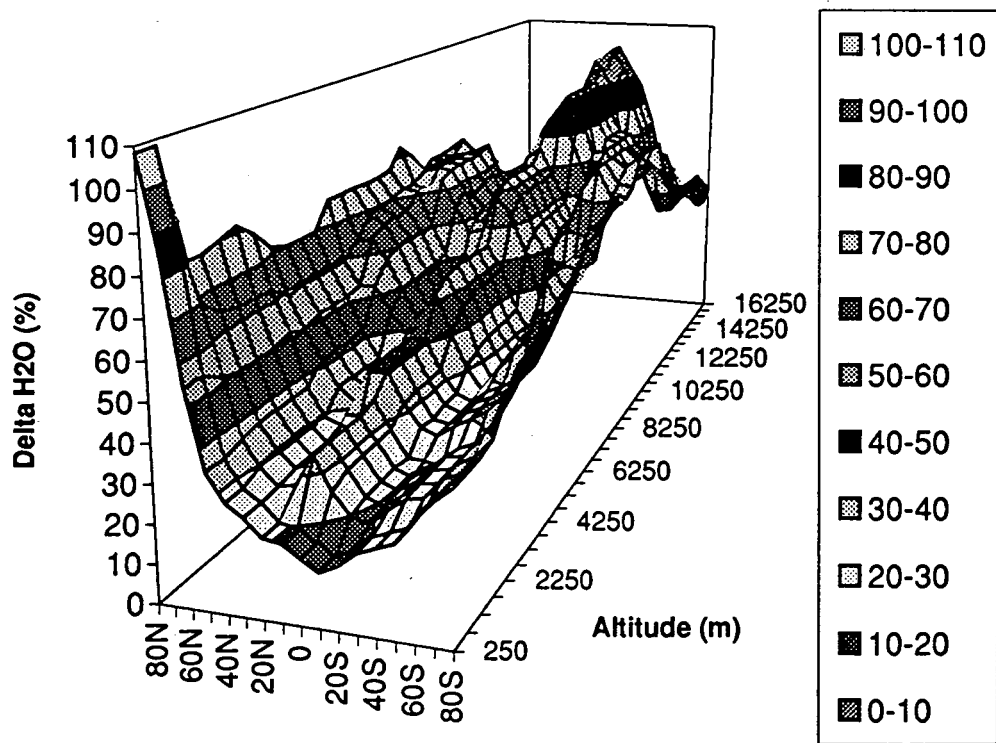


Figure 11. Calculated percentage changes in tropospheric water vapour between 2050 and 1990 as function of latitude and altitude.

Three runs with the tropospheric chemistry/transport model were done in addition to the reference run. First, only the temperatures were increased by the amount calculated by Wang et al. (1992). Then, only the water vapour fields were changed, and finally both temperatures and water vapour were changed simultaneously. In all cases the model was run for 50 years to reach steady state. The calculated changes in tropospheric annual averages of O<sub>3</sub>, OH, CO and CH<sub>4</sub> are given in Table 1.

*Table 1. Changes in tropospheric annual mean levels of O<sub>3</sub>, OH, CO and CH<sub>4</sub> in response to increased temperatures and water vapour densities.*

Case	O <sub>3</sub>	OH	CO	CH <sub>4</sub>
delta T	- 2.4 %	- 1.8 %	1.0 %	- 7.2 %
delta H <sub>2</sub> O	- 8.0 %	19.6 %	- 10.4 %	- 11.1 %
delta T and delta H <sub>2</sub> O	- 10.2 %	17.1 %	- 9.5 %	- 17.6 %

In the combined case (delta T and delta H<sub>2</sub>O) ozone reductions are calculated in the entire model domain for all months. The reductions are usually largest around Equator at 4-5 km where they reach 20-22 %. At 10-12 km the reductions are in the range 6-14 % depending on month and latitude. Figure 12 shows the calculated changes in tropospheric O<sub>3</sub> for the combined case for January. In addition to the large reductions at Equator, there are also significant reductions at low altitudes over high Southern latitudes. Strong reductions over high Southern latitudes are also calculated for December and February. Such marked reductions are not so pronounced at high Northern latitudes in June, July and August. The levels of NO<sub>x</sub> and other ozone precursors (CO and hydrocarbons) are higher at these latitudes, and thus the ozone producing reactions are more important for the ozone budget.

Figure 13 shows the calculated changes in OH for January. The highest percentage increases in OH are found at high Northern latitudes, while the increases are smallest in the middle and lower troposphere at lower latitudes. In May, June and July, the largest percentage OH increases are found at high Southern latitudes.

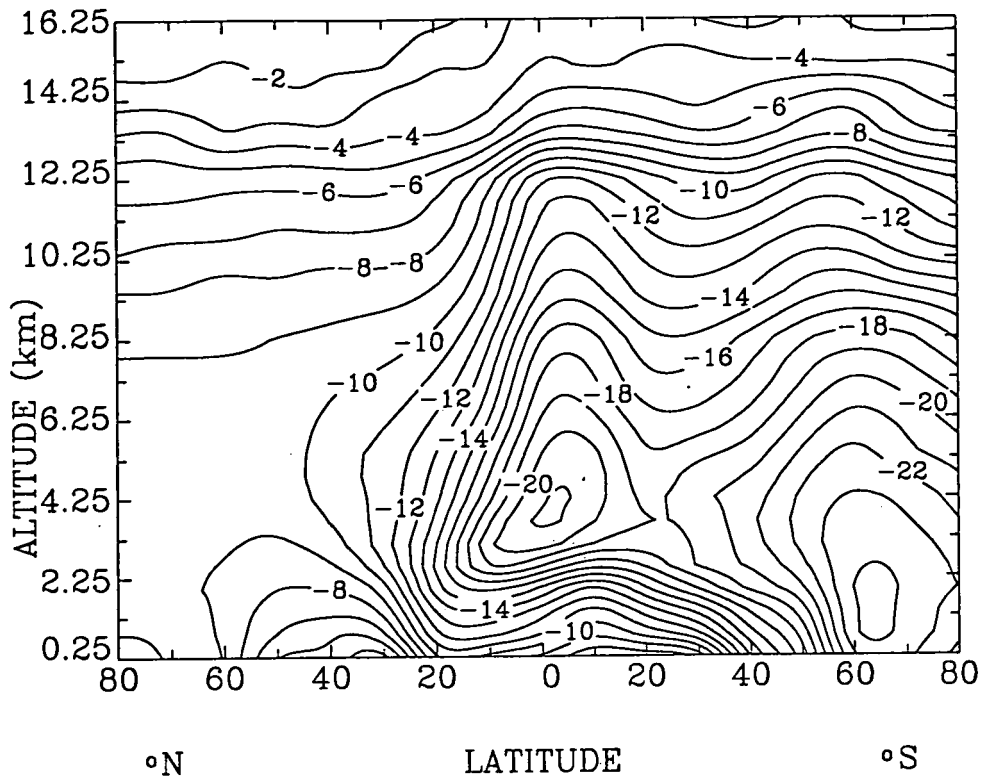


Figure 12. Calculated percentage changes in tropospheric  $O_3$  for January in response to increased temperatures and water vapour levels.

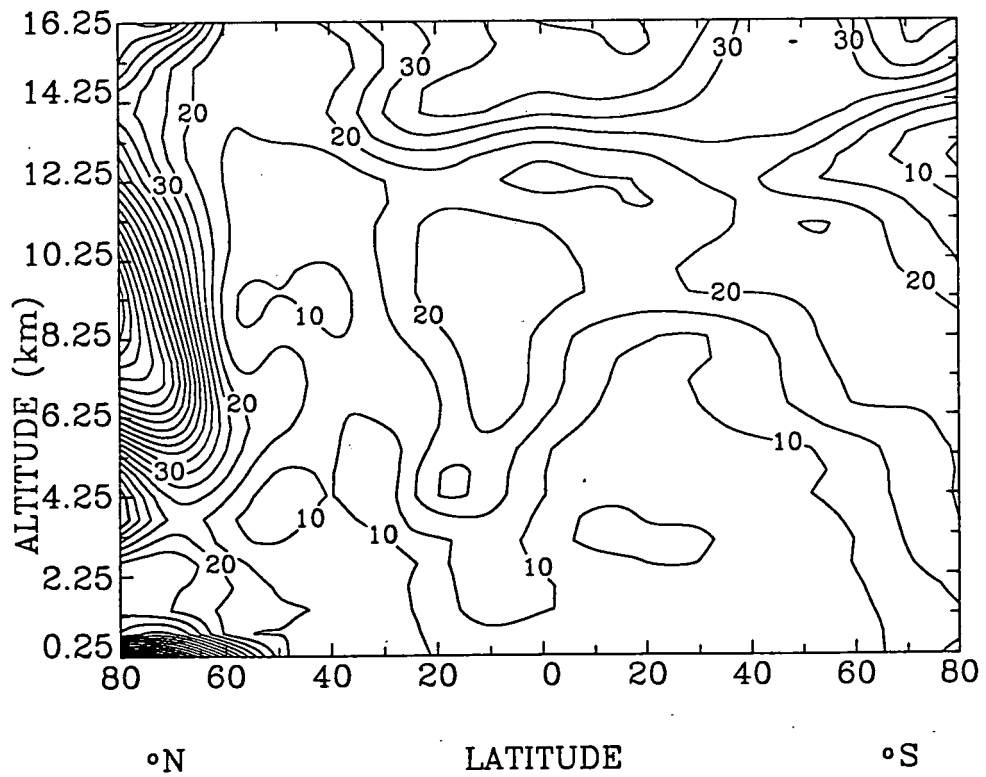


Figure 13. Calculated percentage changes in OH for January in response to increased temperatures and water vapour levels.

### 3. Discussion and conclusions

Solar UV-B fluxes are to a large extent determined by the thickness of the overlying ozone column. The changes in total ozone column will therefore mainly affect the J-values in the UV-B region. In terms of global averages, the largest changes are calculated for the dissociation of ozone that yields O(<sup>1</sup>D),  $J_B(O_3)$  (see figure 1 and 2), while the J-values for HNO<sub>3</sub>, H<sub>2</sub>O<sub>2</sub> and HCHO change by 1-7% between 1979 and 1993. Increases in  $J_B(O_3)$  will intensify the rate at which OH radicals are produced through R1 and R2. Due to damping mechanisms in the chemistry, however, the increase in the concentrations of OH are less than the increase in the production rate of OH (Fuglestedt et al., 1994). Higher levels of OH will enhance the levels of HO<sub>2</sub>, which will further increase the loss of ozone through the reaction

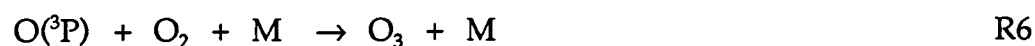
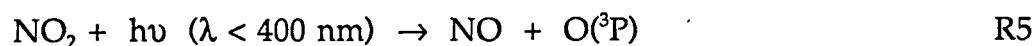


The loss of O<sub>3</sub> through R1 and R2 will also contribute to reduced levels of O<sub>3</sub> if the NO<sub>x</sub> levels are low.

The ozone increases calculated for April at middle and high Northern latitudes are due to the high levels of ozone precursors that have accumulated during the winter. The chemical activity intensify in response to the increases in UV radiation. In regions where NO<sub>x</sub> is abundant, the higher levels of HO<sub>2</sub> increase the rate of R4 which is a key reaction in the production of ozone:



The NO<sub>2</sub> produced will photolyze and produce ozone through the reactions



Comparison of our results with observations indicate that the reductions in stratospheric ozone may already have had impacts on tropospheric chemistry. Schnell et al. (1991) found a 17 % reduction in surface ozone at the South Pole in December, January and February over the period 1976-1990. Schnell et al. discuss several explanations and suggest that increased photochemical destruction of O<sub>3</sub> from increased UV penetration is a principal mechanism. Our calculations give reductions in Antarctic tropospheric ozone of the same magnitude between December 1979 and December 1993 (figure 5).

The growth rate of methane has also showed significant reductions during recent years. The rate of increase was about 20 ppbv/yr in the late 1970s, 17-21 ppbv/yr

during 1978-82, 10 ppb/yr in 1990 and about 5 ppbv/yr in 1992 (Blake and Rowland, 1988; WMO, 1992; Steele et al., 1992; Dlugokencky et al., 1994). The reduction in growth rate was strongest in the Northern hemisphere. There are several possible explanations for this development. Increase in OH due to increased UV radiation may have contributed to the observed reductions. Results from Fuglestvedt et al. (1994), based on *modelled* changes in total ozone, indicate that this mechanism may have contributed by about 1/3 of the observed reduction in the growth rate between 1980 and 1990. The present results, based on *observed* total ozone trends from June 1979 through May 1994, give reductions in the growth rate of 4 ppb/yr for 1992 and almost 7 ppbv/yr for 1993 due to changing UV fluxes. In our calculations the growth rate reductions are of similar magnitude in both hemispheres. This is not the case in the observations by Dlugokencky et al. (1994) who find much larger reductions in the Northern than in the Southern hemisphere. It may thus be concluded that increased UV radiation alone can not explain the development in the growth rate of methane, but that this mechanism may have contributed to the observed reduction. Based on the growth rate figures given above, the studied mechanism may have contributed by 1/4 - 1/3 of the observed reductions from 1979 through 1992. Reduced emissions at higher Northern latitudes is another plausible explanation.

Observations by Novelli et al. (1994) show that the levels of CO dropped by 6.1 %/yr in the Northern Hemisphere and 7.0 %/yr in the Southern Hemisphere between June 1990 and June 1993. Our calculations show (figure 9) that increased UV may reduce CO levels by up to 3.5 % over the period June 1979 through May 1994. The calculations give stronger reductions at high latitudes, while Novelli et al. (1994) report the strongest reductions in the tropics. This indicates that increased UV can not explain the CO trend, although it may have contributed.

Changes in climate parameters as temperature and water vapour give rise to a complex set of responses in tropospheric chemistry. Most of the reaction rate constants are temperature dependent; some are increasing with temperature, while other are decreasing. The degree of temperature dependence also shows large variations. In the model run where the temperatures were increased (the delta T case) the tropospheric annual level of OH was reduced, while the level of CH<sub>4</sub> was also reduced at the same time. This is due to a larger percentage increase in the reaction rate constant for the reaction between OH and CH<sub>4</sub> than the percentage reduction in OH levels. For instance, an increase from 288 to 292 K increase the reaction rate by 9 %. The modelled OH reductions in the regions of importance for methane oxidation (the lower tropical troposphere) are typically about 2 %. The net effect is an overall enhancement of the loss, given by  $k \cdot [\text{OH}]$ , and thus reduced methane concentrations. However, OH increases are also found in limited regions, mainly at middle and high latitudes. The chemical production of CO is increased since CO is formed in the degradation of methane. For other gases the net effect is not as obvious as for CH<sub>4</sub> and CO. For instance, the levels of OH are controlled by a number of reactions with

different temperature dependencies.

In the delta T case, the modelled changes in temperature from Wang et al. (1992) were added to the reference temperatures used in the tropospheric chemistry/transport model. Due to the non-linear relations between the reactions rate constant and temperature for most of the reactions, the results will also depend on the reference levels, and not only on the magnitudes of the temperatures changes. This will introduce some uncertainty to the obtained results.

In the delta H<sub>2</sub>O case, the OH levels are increased due to higher production through R1 and R2. As in the case of increased OH production from increased UV, the OH perturbations are somewhat damped by the chemical interactions in the troposphere. The increase in OH levels enhance the levels of HO<sub>2</sub>, thereby increasing the loss of ozone through R3. In regions with low levels of NO<sub>x</sub>, there will also be increased loss of O<sub>3</sub> through R1 and R2. The increases in OH will further reduce the levels of CO and CH<sub>4</sub>.

In terms of global annual averages, the results from the delta T and the delta H<sub>2</sub>O cases are nearly additive. Relatively large reductions in O<sub>3</sub> and CH<sub>4</sub> are calculated for the combined case, about 10 % and 18 %, respectively.

Several studies have emphasized the importance of sulphate particles in the context of climate change (e.g. Charlson et al., 1991; Taylor and Penner, 1994). Sulphate is formed when SO<sub>2</sub> is oxidized either in the gas phase by OH or in the liquid phase where O<sub>3</sub> and in particular H<sub>2</sub>O<sub>2</sub> are important oxidizing species. We have shown that increased UV radiation, temperatures and water vapour densities increase OH and generally reduce tropospheric O<sub>3</sub>. In addition, both increased UV radiation as well as increased temperatures and water vapour densities give higher levels of H<sub>2</sub>O<sub>2</sub> according to our results. Thus, changes in UV, temperatures and water vapour may also affect climate through changes in the sulphur chemistry. Increased levels of OH will also increase the loss of the climate gases HFCs (hydrofluorocarbons) and HCFCs (hydrochlorofluorocarbons).

Our calculations show that reductions in stratospheric ozone reduce the levels of important climate gases as tropospheric ozone and methane via UV changes. Thus, this constitutes a negative indirect effect of the ozone depleting gases on climate change. Furthermore, our study indicate that a global warming of 3.88 K will significantly reduce the levels of tropospheric O<sub>3</sub> and CH<sub>4</sub>, which constitutes a negative feedback in the climate system.

In the studies of the response in tropospheric chemistry to changes in stratospheric ozone, only the changes in UV radiation are considered, while the fluxes of ozone from the stratosphere to the troposphere are kept constant. The validity of the results depends critically on the quality of the total ozone column data that are used in the calculations of the photolysis rates. Furthermore, we have only studied the effects of changes in two climate parameters, temperature and water vapour. Climate change will initiate changes in several factors that will influence atmospheric



chemistry significantly; for instance occurrence and properties of clouds, emission rates, dynamics, etc. (See Fuglestedt et al. (1994) for a discussion of uncertainties and limitations).

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