# A model calculation of the ozone response to the increase

# in the atmospheric emission of several gases

## $(CO_2, CH_4, N_2O$ and CFCs)

A preliminary Report

by

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## 1. INTRODUCTION

A series of time-dependent, multiple perturbation calculations have been carried out using a 1-D chemical radiative model to describe the response of the ozone layer to the injection in the atmosphere of gases such as the chlorofluorocarbons (F-11 and F-12), $CH_3CCl_3$, $CCl_4$, $CO_2$, $CH_4$ and $N_2O$. The effect of nitric oxide released by the engines of aircraft is also considered in some model cases.

The purpose of this preliminary note is to briefly summarize the results obtained by this model. A number of graphs with the main model output is given hereafter. Full details of the calculations will be published in scientific journals.

The model which is used in the present work is one-dimensional and extends from the earth's surface to 100 km altitude. The chemical routine considers a number of species belonging to the $O_x$, $HO_x$, $NO_x$ $ClO_x$ and $CO_x$ families. The chemical rate constants are taken from the JPL publication 1983. The vertical profile of these species is calculated from continuity equations in which the vertical flux is determined for a prescribed "eddy diffusion coefficient". The temperature which determines the local value of most reaction rates is self-consistently calculated by a radiative method which considers the absorption of the solar UV radiation by $O_3$ and $O_2$ and the infra-red transfer of terrestrial and atmospheric radiation by $CO_2$, $O_3$ and $H_2O$. A 50 percent cloud coverage is assumed, the top of the clouds being located at 5 km altitude.

## 2. EVOLUTION OF THE TRACE GASES AMOUNT IN THE PAST (1940-1983)

The following mixing ratios (f) or world emissions (E) with the corresponding references have been adopted as a function of time (t) in the model calculations.

$CO_2$ mixing ratio :

$1940 \leqq t \leqq 1957$     $f = 2.7 \times 10^{-4} \exp[.00141(t-1849)]$        (1)

$1957 < t \leqq 1980$     $f = 2.7 \times 10^{-4} + 4.44 \times 10^{-5} \exp[.019(t-1957)]$   (1)

$1980 < t \leqq 1983$     $f = f(1980) \times \exp[.005(t - 1980)]$        (2)


$N_2O$ tropospheric mixing ratio :

$1940 \leqq t \leqq 1980$     $f = 2.85 \times 10^{-7} + .14 \times 10^{-7} \exp[.04(t - 1977)]$   (1)

$1980 < t \leqq 1983$     $f = f(1980) \times \exp[.002(t - 1980)]$        (2)


$CH_4$ tropospheric mixing ratio :

$1940 \leqq t \leqq 1980$     $f = 10^{-6} + .65 \times 10^{-6} \exp[.035(t - 1979)]$     (1)

$1980 < t \leqq 1983$     $f = f(1980) \times \exp[.01(t - 1980)]$        (2)


$CH_3CCl_3$ world emission :

$1940 \leqq t \leqq 1950$     $E = 0$

$1950 < t \leqq 1976$     estimates from (3)

$1976 < t \leqq 1980$     linear with time

$1980 < t \leqq 1983$     $E = 500$ kt/year        (2)


$CCl_4$ world emission :

$1940 \leqq t \leqq 1980$     estimates from (4)

$1980 < t \leqq 1983$     $E = 100$ kt/year        (2)


CFC 11 and 12 world emission :

CMA values (see figure)

(1)    D.J. Wuebbles, M.C. Mac Cracken and F.M. Luther, A proposed Reference Set of Scenarios for Radiatively Active Atmospheric Constituents, Prepared for the United States Department of Energy, DOE/NBB-0066, October 1984.

(2)    Working value (C.M.A).

(3)    R.G. Prinn et al., The Atmospheric Lifetime Experiment. 5. Results for $CH_3CCl_3$ based on three years of data, J.G.R., 88, c13, 8415-8426, 1983.

(4)    P.G. Simmonds et al., The A.L. Experiment. 6. Results for Carbon Tetrachloride based on 3 years data, J.G.R., 88, c13, 8427-8441, 1983 (After 1958 : lower limit).

## 3. SCENARIOS FOR THE FUTURE    (1983-2040)

Four scenarios running through to 2040 have been considered, which have certain assumptions about trends in non-CFC gases in common, but which differ in the CFC emission patterns.

The assumptions about the non-CFC gases are as follows :

a)    $CO_2$ - 0.5%/year exponential growth (as specified in the 1984 NAS report) (fig. 1) $f = 340 \times 10^{-6} \times \exp[0.005(t - 1980)]$

b)    $N_2O$ - 0.2%/year exponential growth. (fig. 2). $f = 301 \times 10^{-9} \times \exp[0.002(t - 1980)]$

c)    $CH_4$ - 1%/year exponential growth (fig. 3) - this is at the conservative end of the generally accepted 1-2% year range. $f = 1.67 \times 10^{-6} \times \exp[0.01(t - 1980)]$

d)    $CH_3CCl_3$ - Constant emission at 500kt/year (fig. 4).

e)    $CCl_4$ - Constant emission of 100kt/year (fig. 4).

The CFC emissions for the four scenarios are shown graphically in figures 5 and 6; they are derived from the following productions :

1.    Constant production on the basis of the year 1983 - the base line case.

2.    Productions grow by 3% year until they reach 1.5 times current productions in 1997. This simulates the extension of the EEC production cap provision to a global scale.

3.    Aerosol usage world-wide phased out over four years, but other uses grow at 3% year until they reach 1.5 times current productions. This simulates the aerosol ban proposed for the CFC protocol, associated with a global capacity cap equivalent to that prevailing in the EEC.

4.    Identical to 3 but with the 3% per year growth allowed to continue indefinitely. This is equivalent to the protocol proposal which does not include a capacity cap.
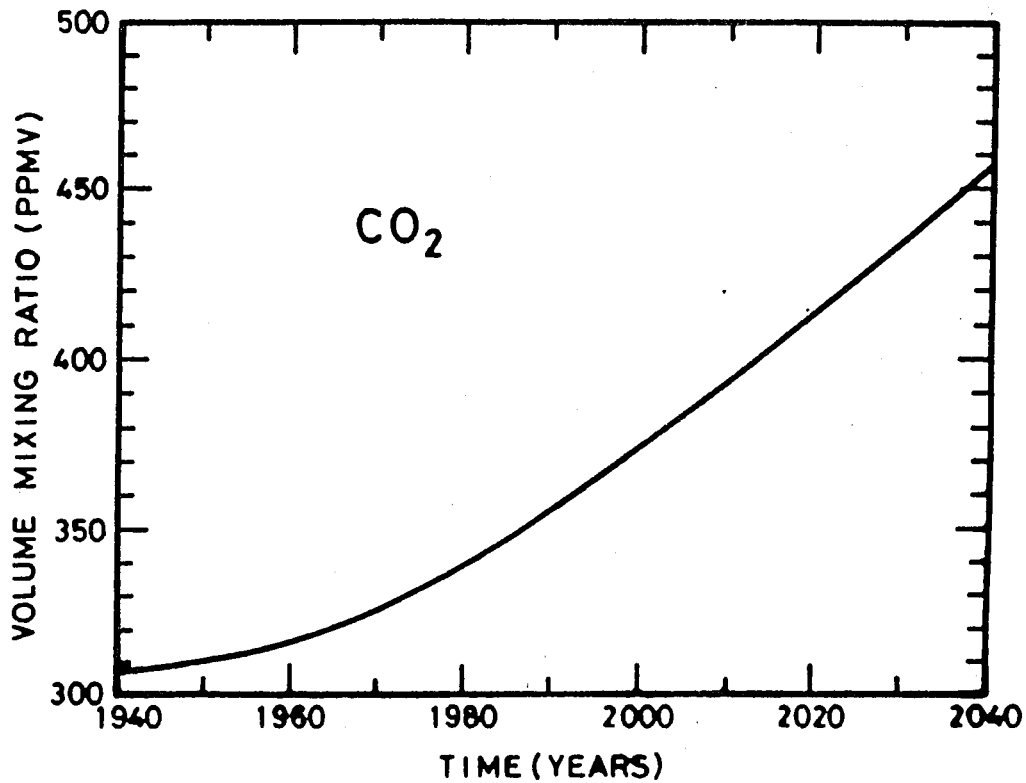
Figure 1 Assumed volume mixing ratio for carbon dioxide as a function of time. Before 1980, the expression suggested by Wuebbles et al. (1984) has been adopted. An increase of 0.5% per year has been assumed afterwards. The value in 1980 is 340 ppmv.
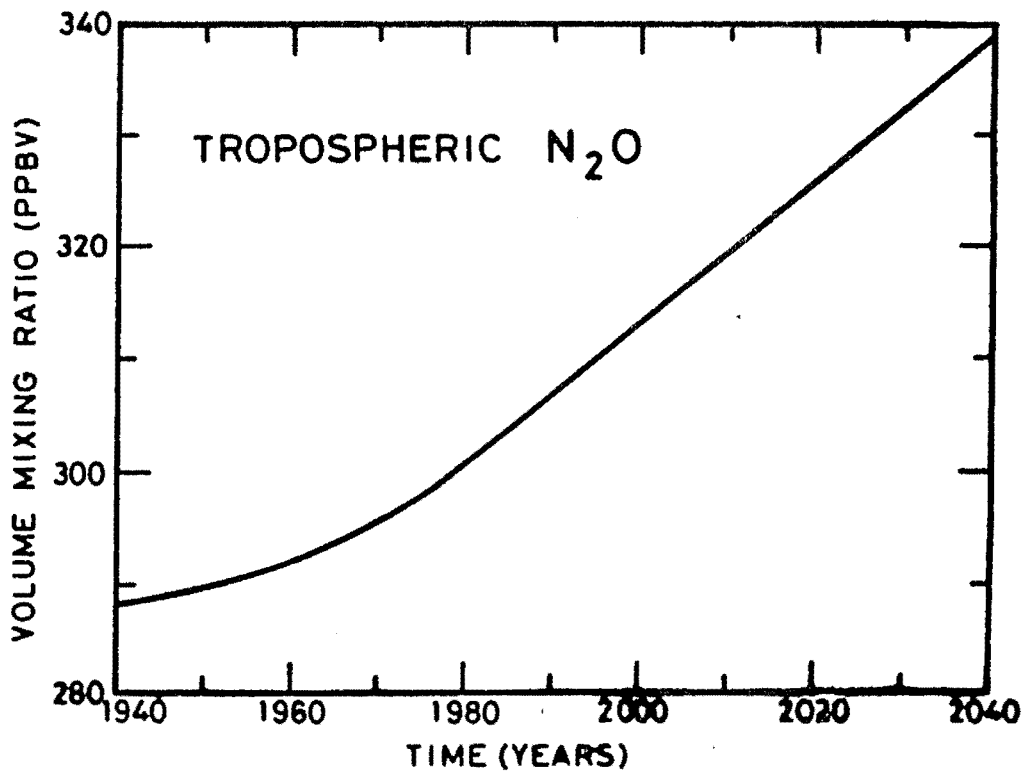
Figure 2  Adopted tropospheric volume mixing ratio for nitrous oxide as
          a function of time. The expression before 1980 is the estimate
          by Wuebbles et al (1984) and leads to a value of 301 ppbv in
          1980. A 0.2% increase per year is assumed after 1980.
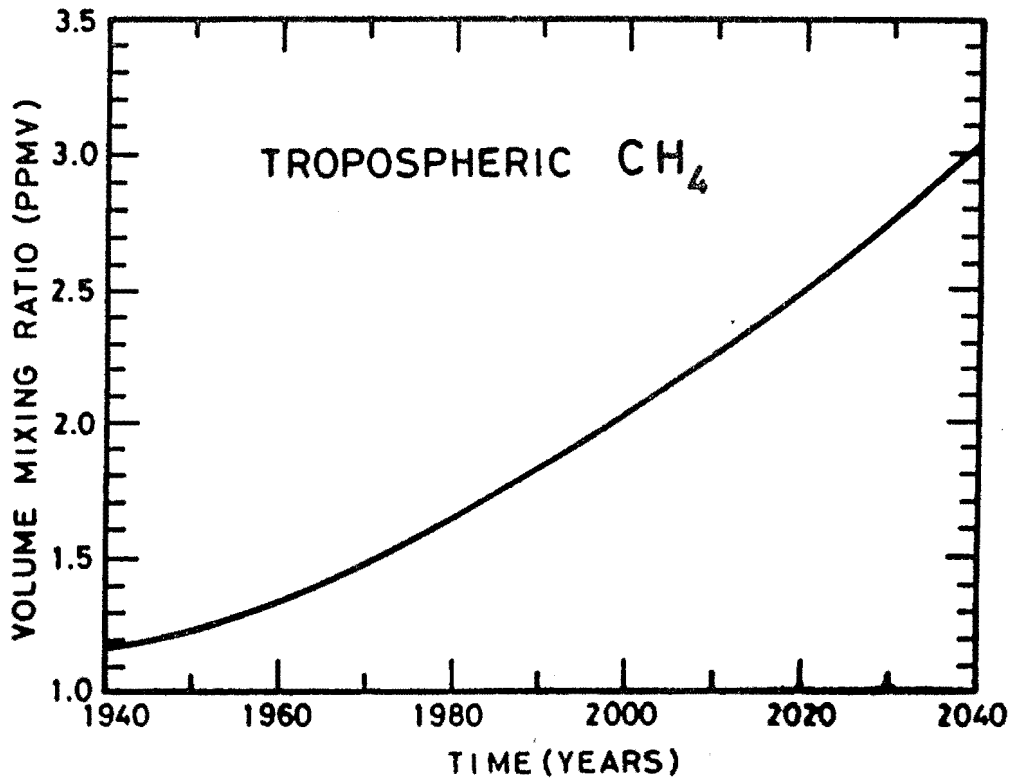
Figure 3   Assumed increase in the tropospheric content in methane. The value before 1980 is taken from Wuebbles et al. (1984) and equals 1.67 ppmv in 1980. A 1% increase per year is adopted after 1980.
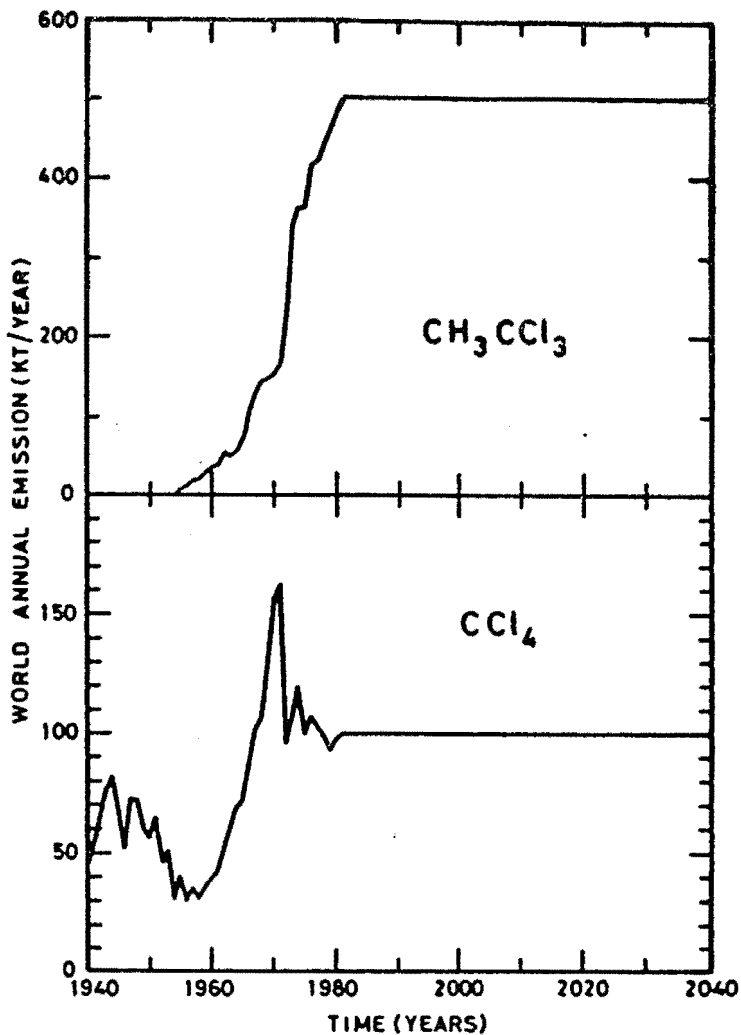
Figure 4 Historical and assumed future emission of methyl chloroform and carbon tetrachloride. Estimates for the past are from the Atmospheric Lifetime Experiment (1983). A constant emission of 500 kt/yr for $CH_3CCl_3$ and 100 kt/yr for $CCl_4$ is adopted after 1980.
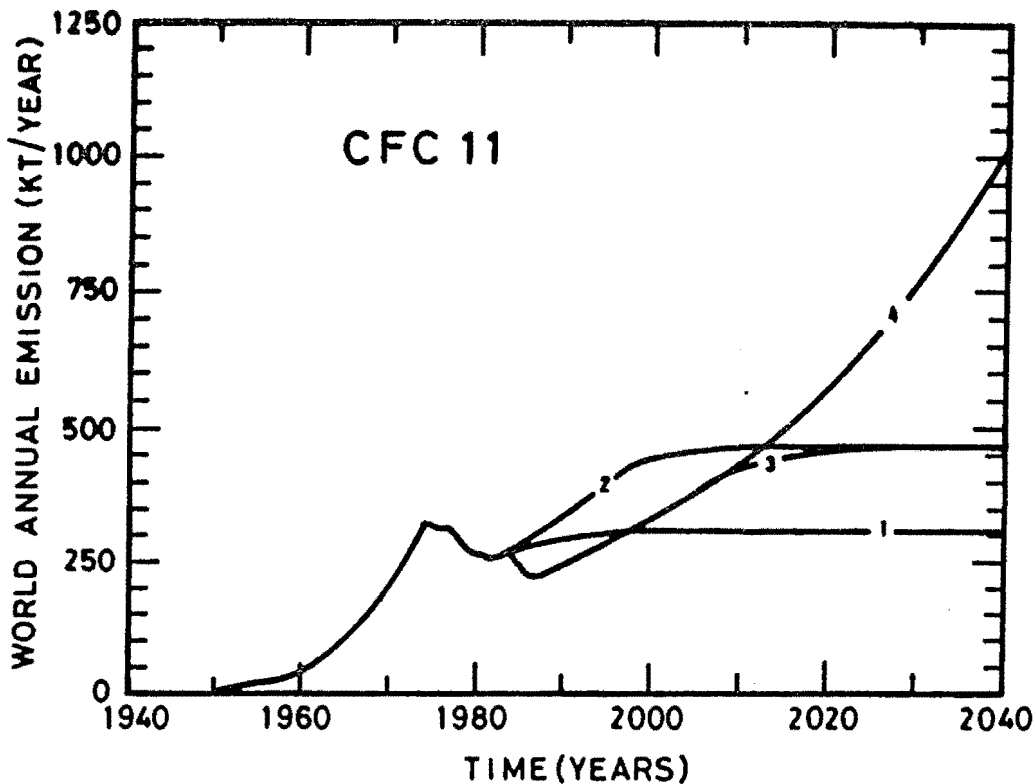
Figure 5    Assumed scenarios for the emission of $CFCl_3$. The past emission (1945 to 1983) is derived from the production estimated by Gamlen et al. (1984). The future emission results from a constant production in scenario 1 and from a 3% yr increase in the production up to the level of 1.5 x its present value (reached in 1997) in scenario 2. Scenario 3 assumes the same increase except in the production of aerosols, which declines between 1983 and 1986; the same capacity cap as in scenario 2 is reached in 2008. Scenario 4 assumes the same conditions as scenario 3 but without any capacity cap.
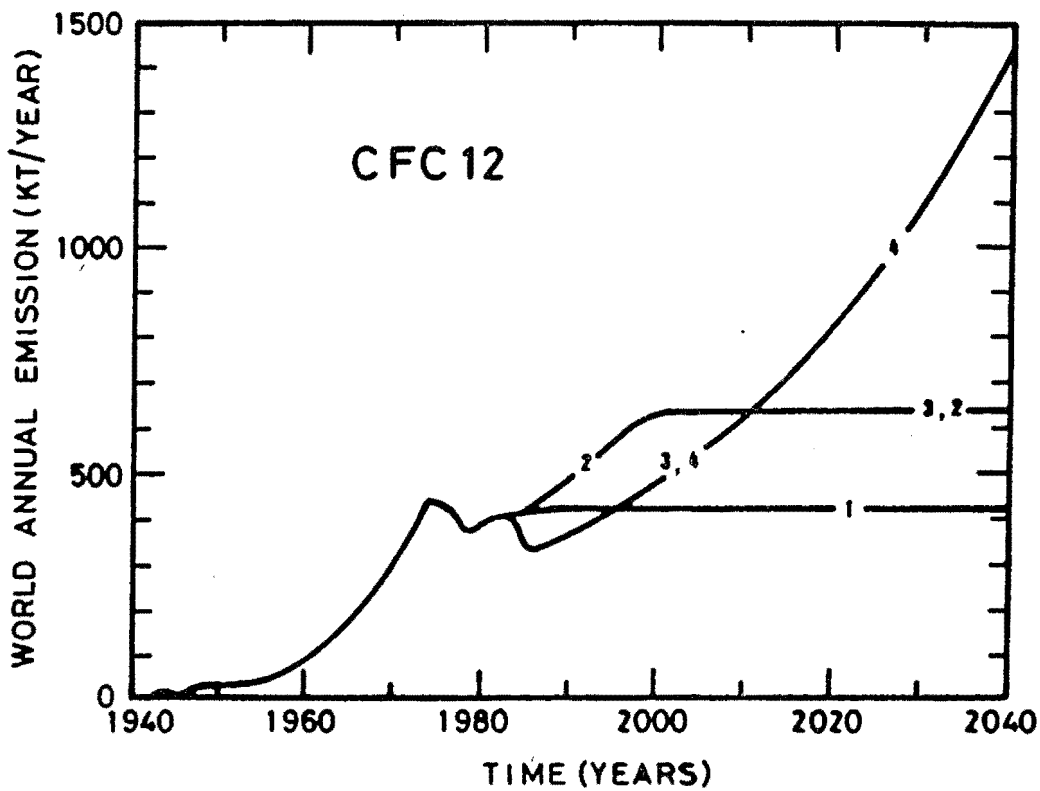
Figure 6   Same as figure 5 but for $CF_2Cl_2$. The decline in the production of aerosols assumed in scenarios 3 and 4 occurs only between 1983 and 1985.

The choice of a 3% growth rate is arbitrary and is intended to demonstrate the calculated effect on stratospheric ozone of future CFC growth. It is not in any sense a prediction that such a growth rate is occurring, or is likely to be sustained in the future. It should be noted that a 3% per annum growth rate up to the year 2040 would result in a five-fold increase in production, and would require around a four-fold increase in installed production capacity.

## NUMERICAL VALUES

### $CFCl_3$ world emission

| | | |
|---|---|---|
| scenario 1 : | $1983 \leqq t \leqq 2002$ | increasing |
| | $2002 < t \leqq 2040$ | E = 309 kt/year |
| scenario 2 : | $1983 \leqq t \leqq 2015$ | increasing |
| | $2015 < t \leqq 2040$ | E = 467.1 kt/year |
| scenario 3 : | $1983 \leqq t \leqq 1987$ | decreasing (219.9 kt/year in 1987) |
| | $1987 < t \leqq 2027$ | increasing |
| | $2027 < t \leqq 2040$ | E = 464 kt/year |
| scenario 4 : | $1983 \leqq t \leqq 1987$ | same as scenario 3 |
| | $1987 < t \leqq 2040$ | increasing (1018.4 kt/y in 2040) |

### $CF_2Cl_2$ word emission

| | | |
|---|---|---|
| scenario 1 : | $1983 \leqq t \leqq 1995$ | increasing |
| | $1995 < t \leqq 2040$ | E = 421.8 kt/year |
| scenario 2 : | $1883 \leqq t \leqq 2009$ | increasing |
| | $2009 < t \leqq 2040$ | E = 638 kt/year |
| scenario 3 : | $1983 \leqq t \leqq 1986$ | decreasing (327 kt/y in 1986) |
| | $1986 < t \leqq 2020$ | increasing |
| | $2020 < t \leqq 2040$ | E = 639.5 kt/year |
| scenario 4 : | $1983 \leqq t \leqq 1986$ | same as scenario 3 |
| | $1986 < t \leqq 2040$ | increasing (1447.3 kt/y in 2040) |

## 4. MODEL RESULTS FOR THE PAST (1940-1983)

Figures 7-13 show results obtained for the historical release and increase of CFCs and other source gases. The predicted present day $Cl_x$ mixing ratio at 50 km is 2.4 ppbv (fig. 7). The change in the temperature at different altitudes is given in fig. 8. The calculated absolute change in the ozone concentration at 40, 30 and 20 km is shown in figures 9-11. Relative variations of the ozone amount at selected altitudes is given in figure 12. The resulting change in the ozone column seems to be characterized by a small increase of 0.2 to 0.3 percent between 1940 and 1983 (fig. 13). This variation results from an ozone depletion in the upper stratosphere and an ozone enhancement below $\sim$ 30 km altitude.

## 5. MODEL RESULTS FOR THE FUTURE (1983-2040)

The following model predictions consider a simultaneous increase in $CO_2$, $N_2O$, $CH_4$ and in the CFC emission. For this last case, the 4 scenarios as described in section 3 have been adopted.

The predicted evolution in the $Cl_x$ mixing ratio at 50 km is shown in fig. 14. A value of the order of 6-8 ppbv is found in year 2040. These numbers are lower than the values obtained in year 2080 when adopting the NASA scenarios (high CFC growth). [NASA report in preparation]. No "high chlorine regime" will be reached (before 2040) by any of the 4 presently adopted scenarios. The change in temperature is given by figures 15 and 16. The relative evolution of the ozone concentration at 50, 40, 30 and 20 km is represented in figures 17-20. One should note that the local behavior of ozone varies significantly with altitude and with the adopted scenario. The absolute and relative change in the ozone concentration as a function of altitude is displayed for scenario 1 in figures 21 and 22 respectively. The change in the ozone column for the 4 scenarios is shown in figure 23. A comparison between
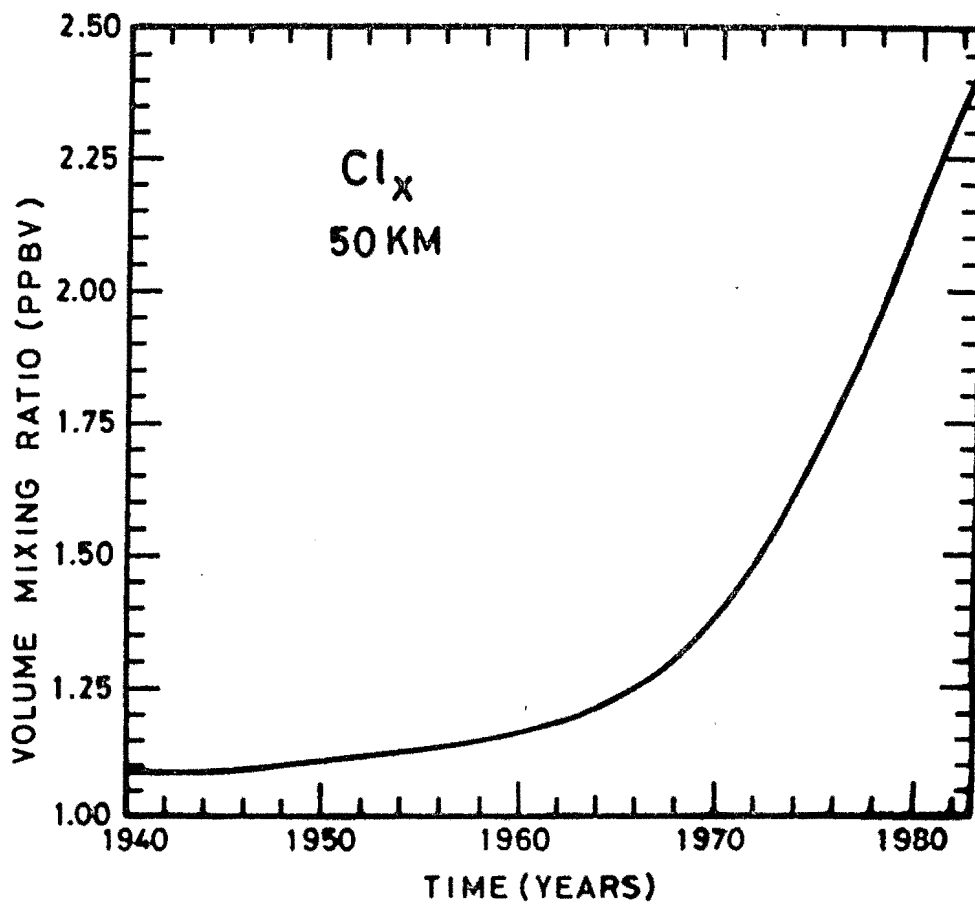
Figure 7  Amount of odd chlorine atoms at 50 km altitude, resulting from
the dissociation of the assumed released chlorocarbons before
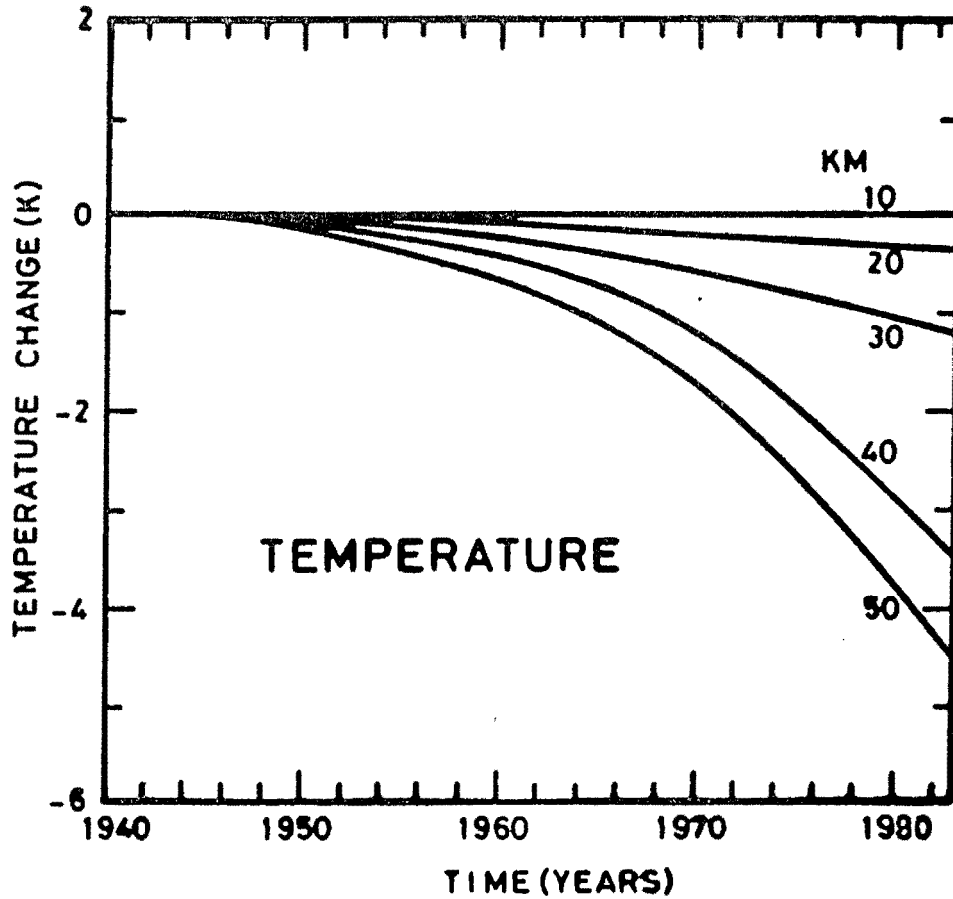1983, as a function of time.

Figure 8   Computed past temperature change from year 1940, as a function of time and at different altitude levels.
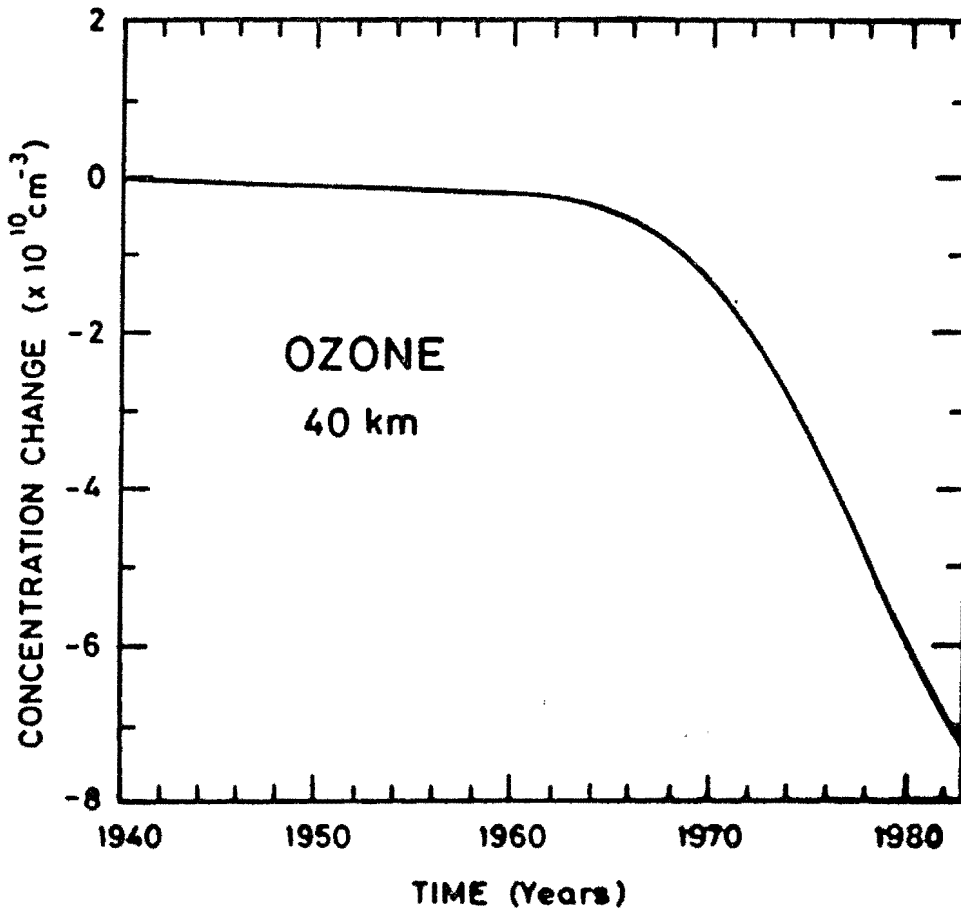
Figure 9  Change in the ozone concentration at 40 km (from its 1940 value) corresponding to the adopted historical scenario, as a function of time.
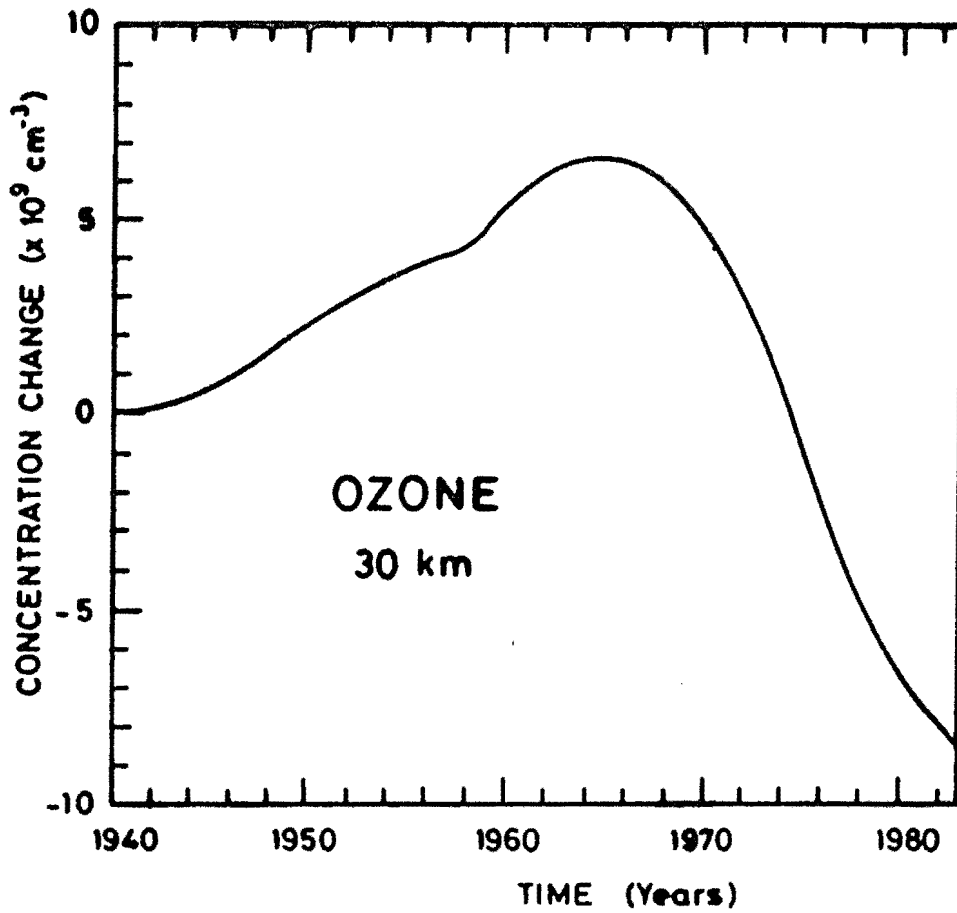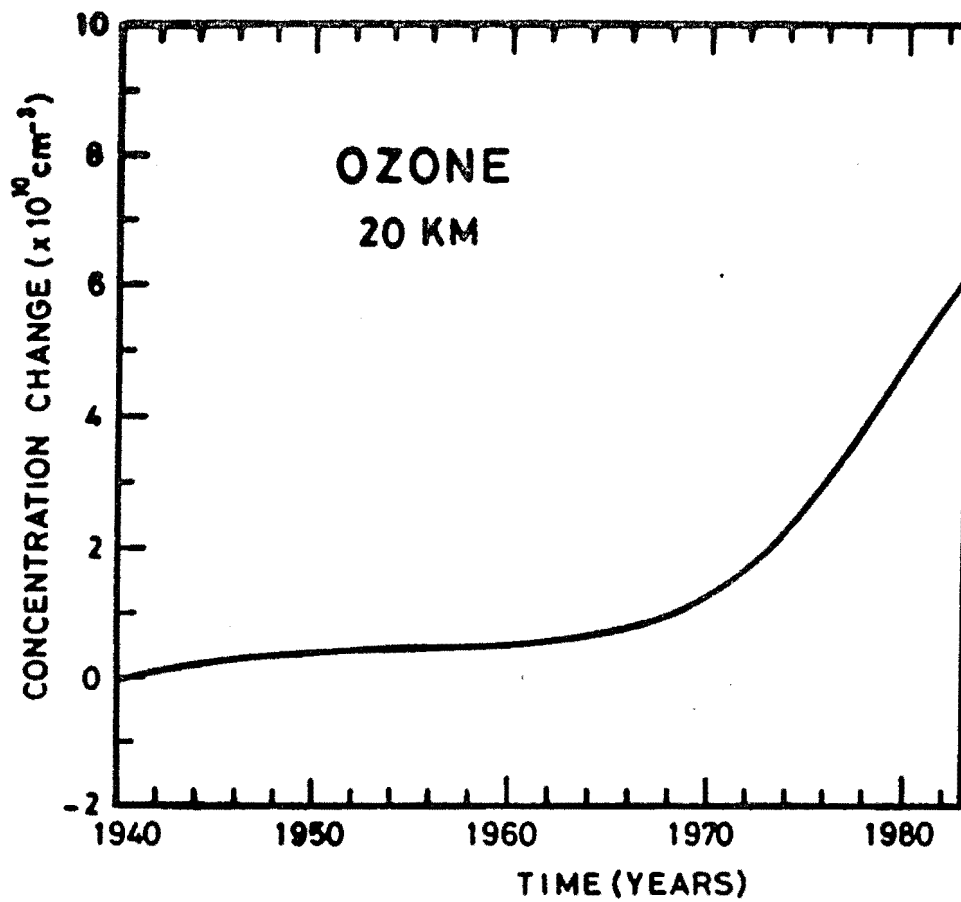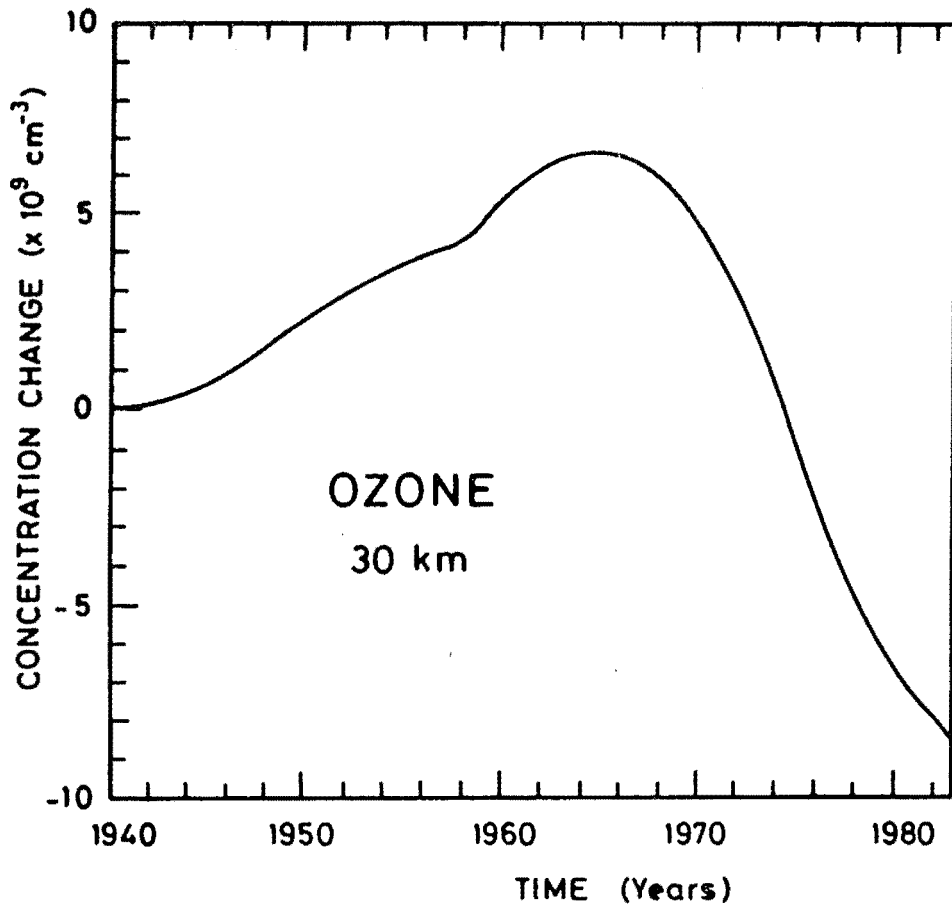
Figure 10 Same as figure 9, but at 30 km altitude.

Figure 11 Same as figure 9, but at 20 km altitude.
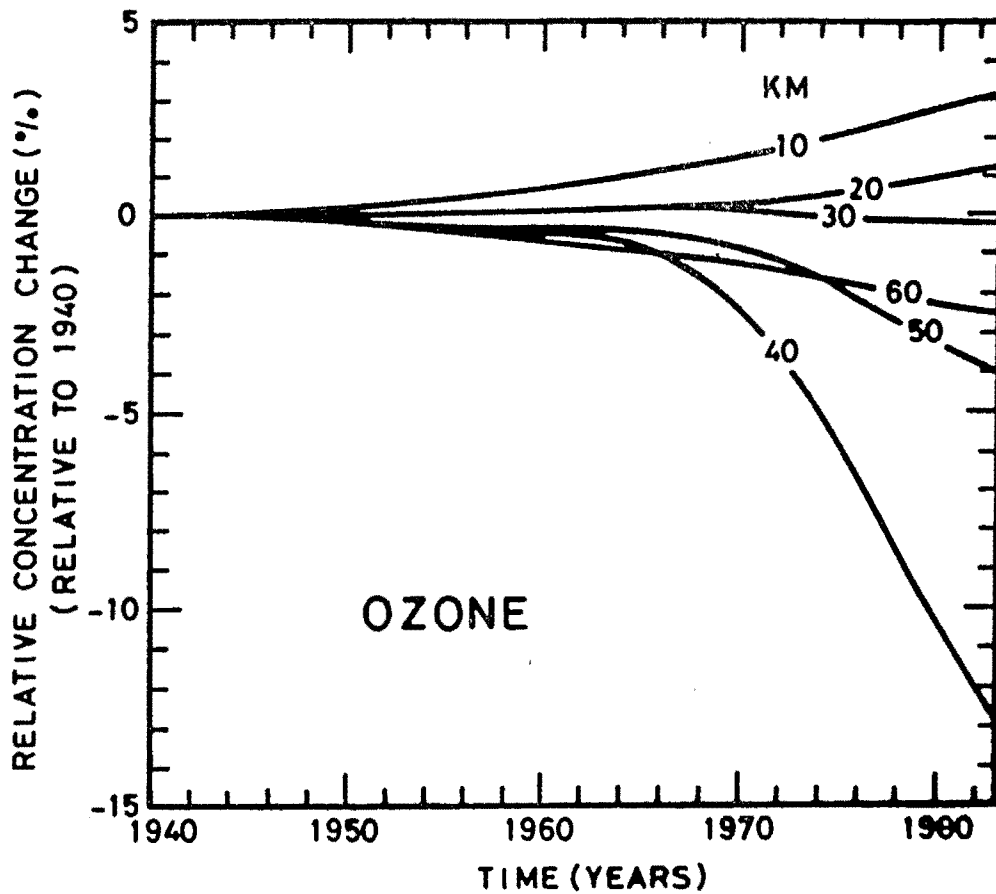
Figure 12 Past relative ozone concentration change computed at different altitude levels.
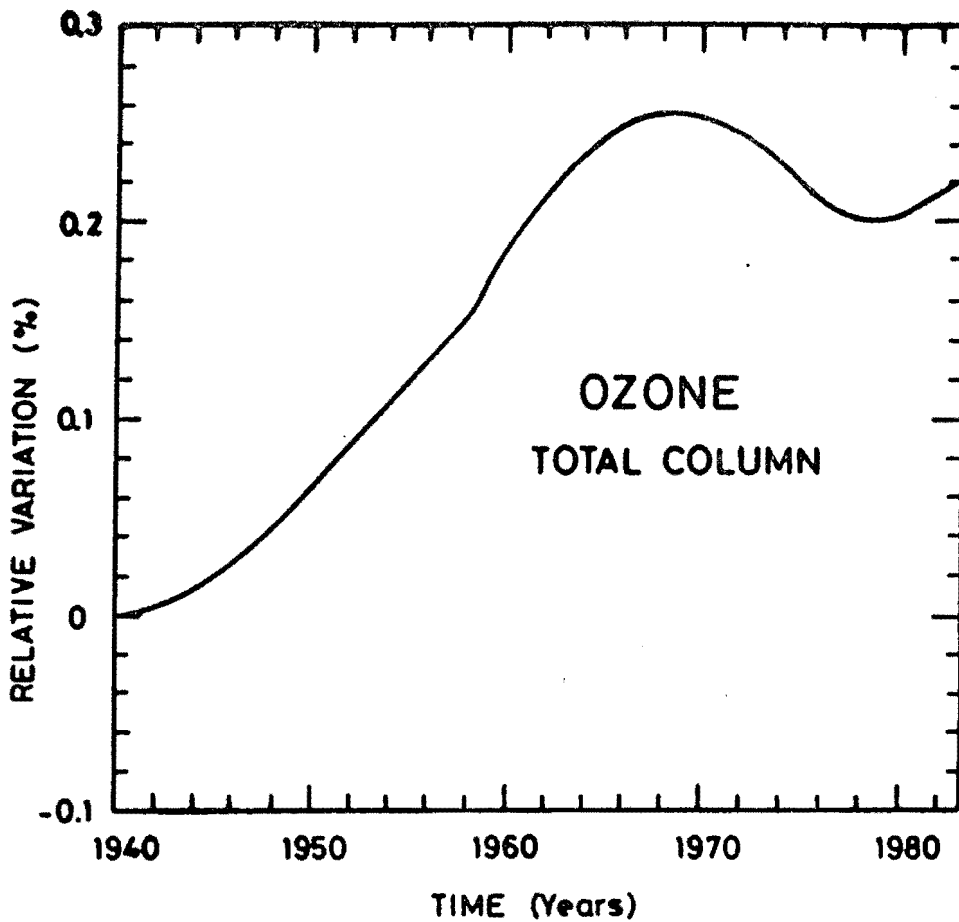
<u>Figure 13</u> Relative variation in the total ozone amount computed for the
past (the initial value being the one of 1940).
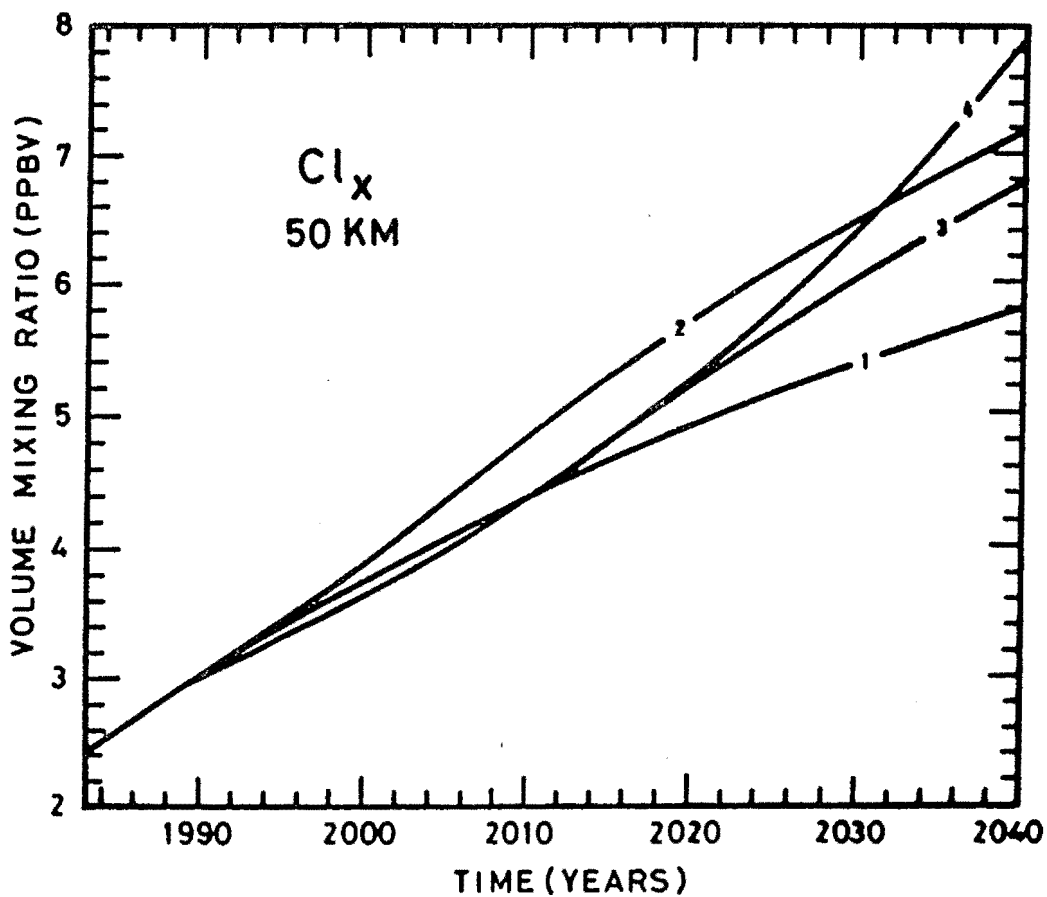
Figure 14 $Cl_x$ volume mixing ratio at 50 km altitude as a function of time, due to the injection of chlorocarbons as assumed in the 4 different scenarios.
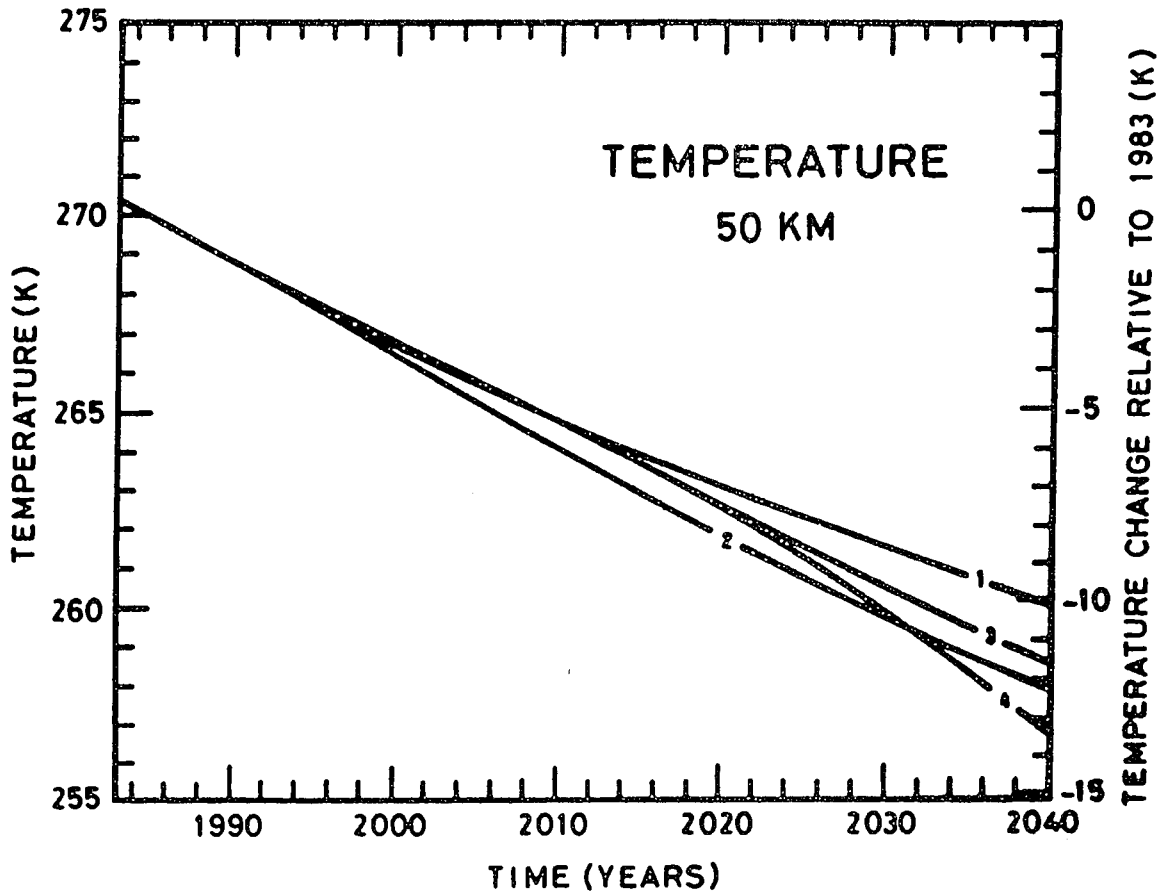
Figure 15 Computed future temperature at 50 km altitude, as a function of time, in the 4 different cases. The right-hand scale indicates its change from 1983.
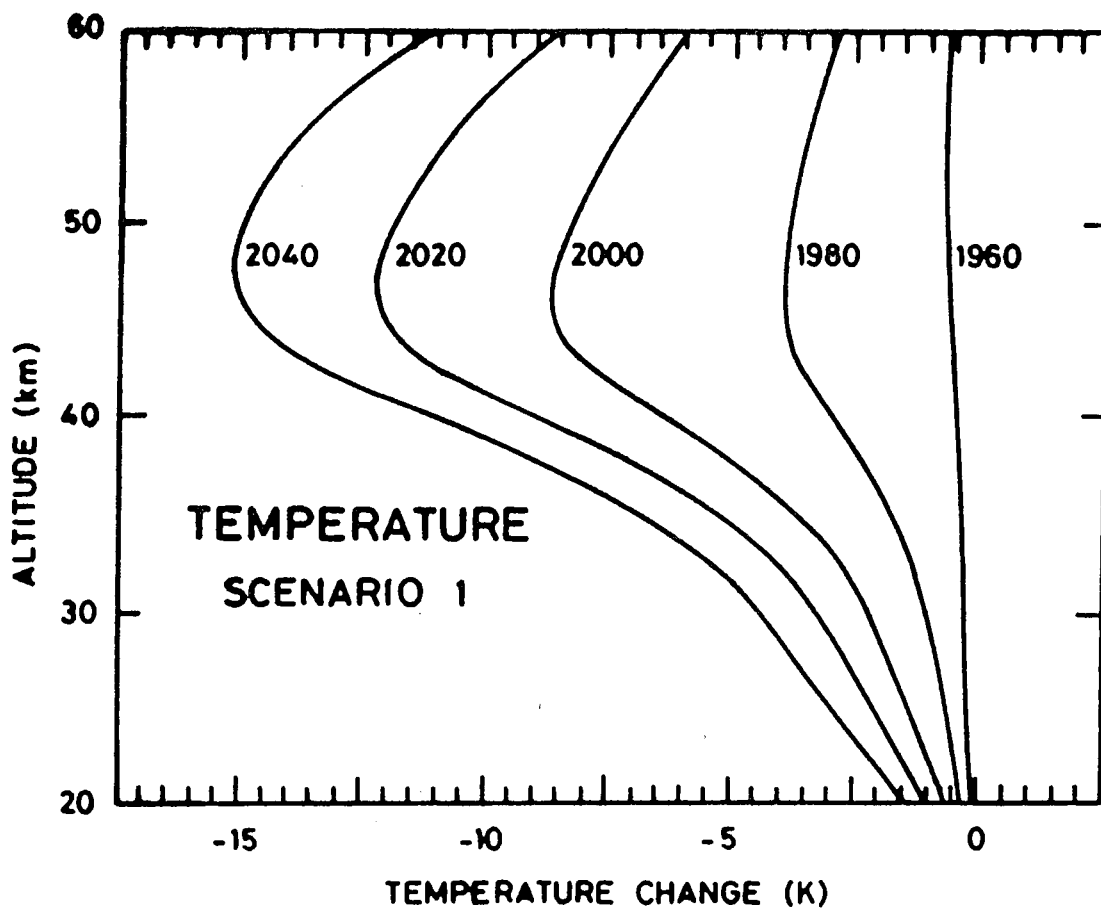
Figure 16 Computed temperature change relative to 1940, in the con-
ditions of scenario 1, as a function of altitude and for
different years.
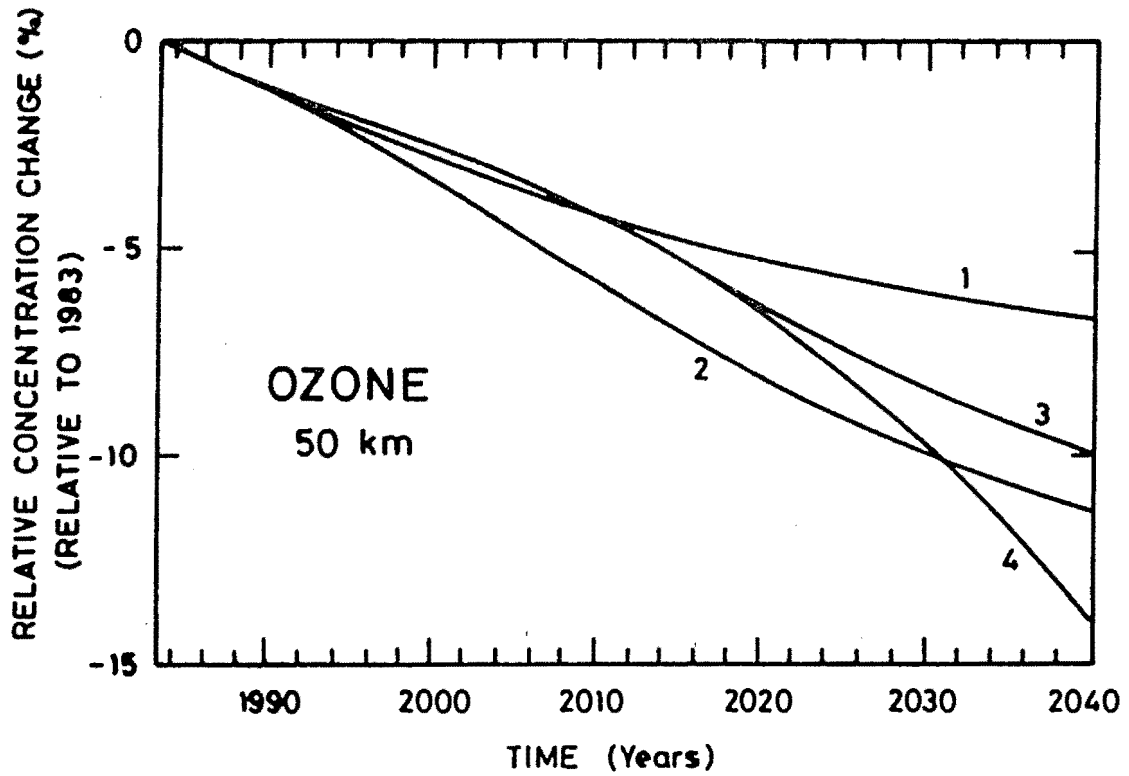
<u>Figure 17</u> Future relative ozone concentration change at 50 km altitude, computed for the 4 different scenarios, as a function of time.
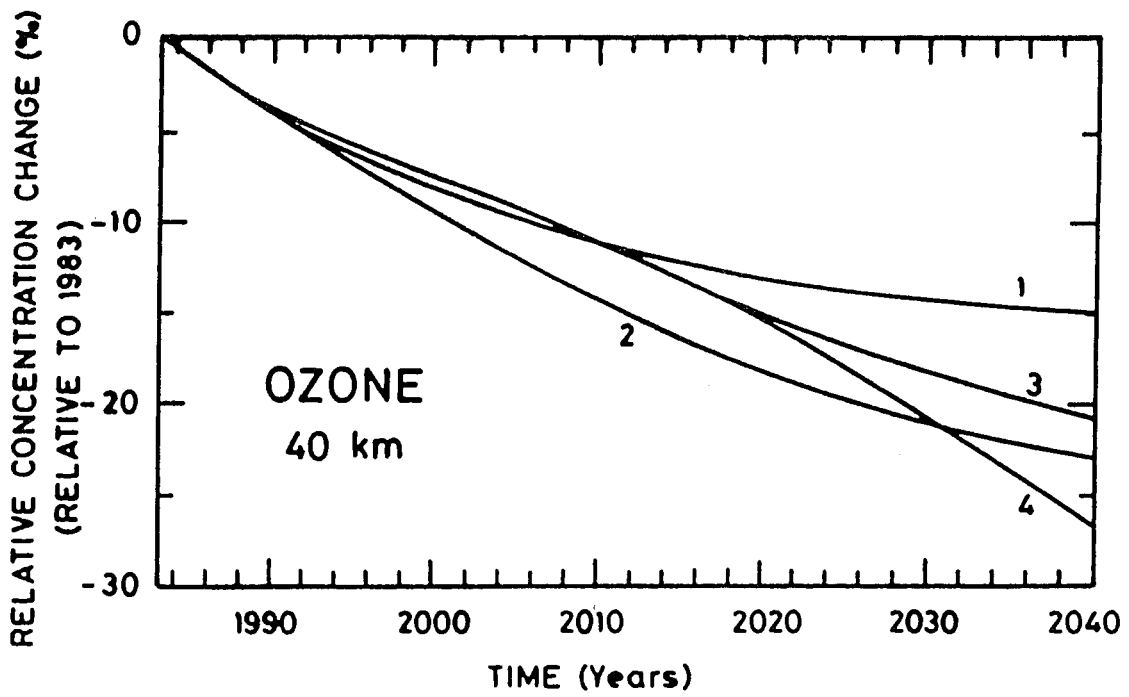
Figure 18 Same as figure 17 but at 40 km altitude.

Figure 19 Same as figure 17 but at 30 km altitude.
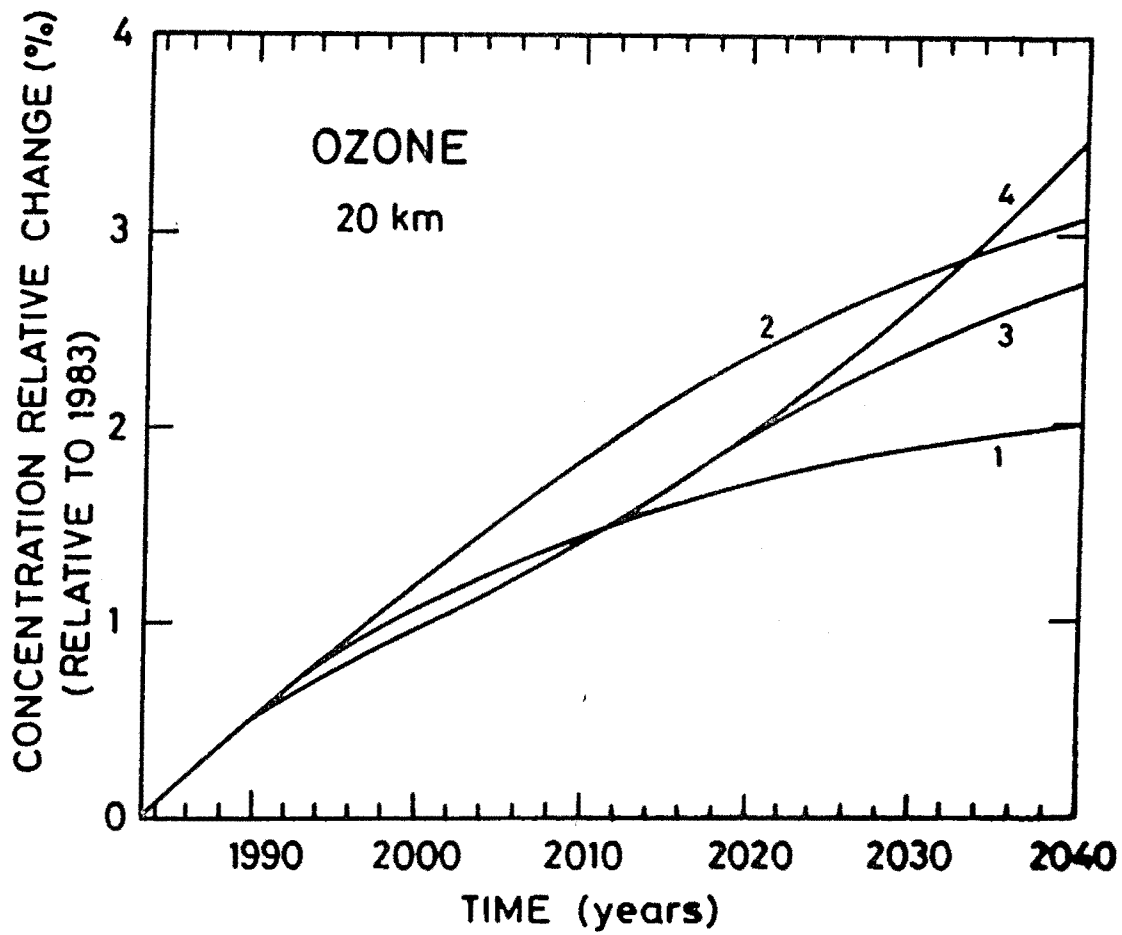
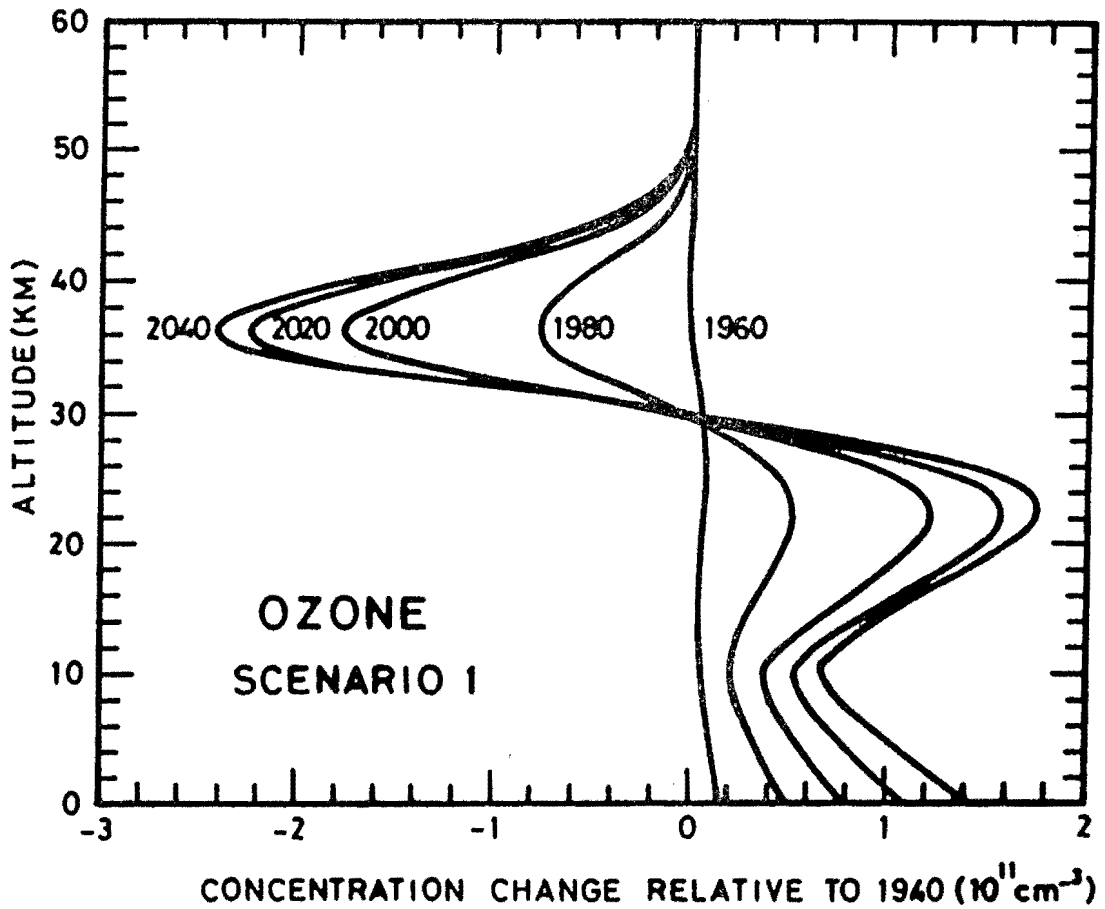Figure 20 Same as figure 17 but at 20 km altitude.

Figure 21 Change in the ozone concentration from its 1940 value, com-
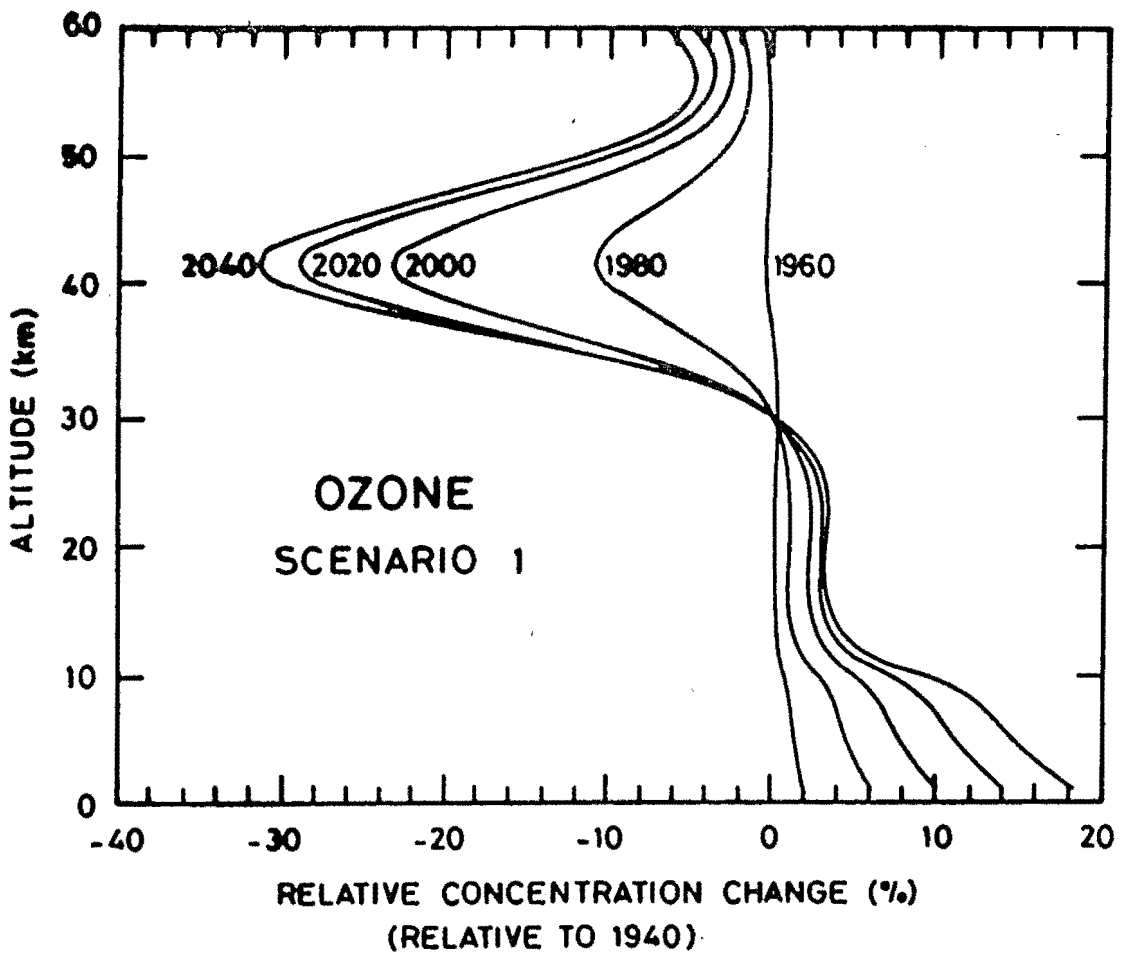puted for scenario 1, as a function of altitude and for
different years.

Figure 22 Same as figure 21, but the ozone change is expressed in re-
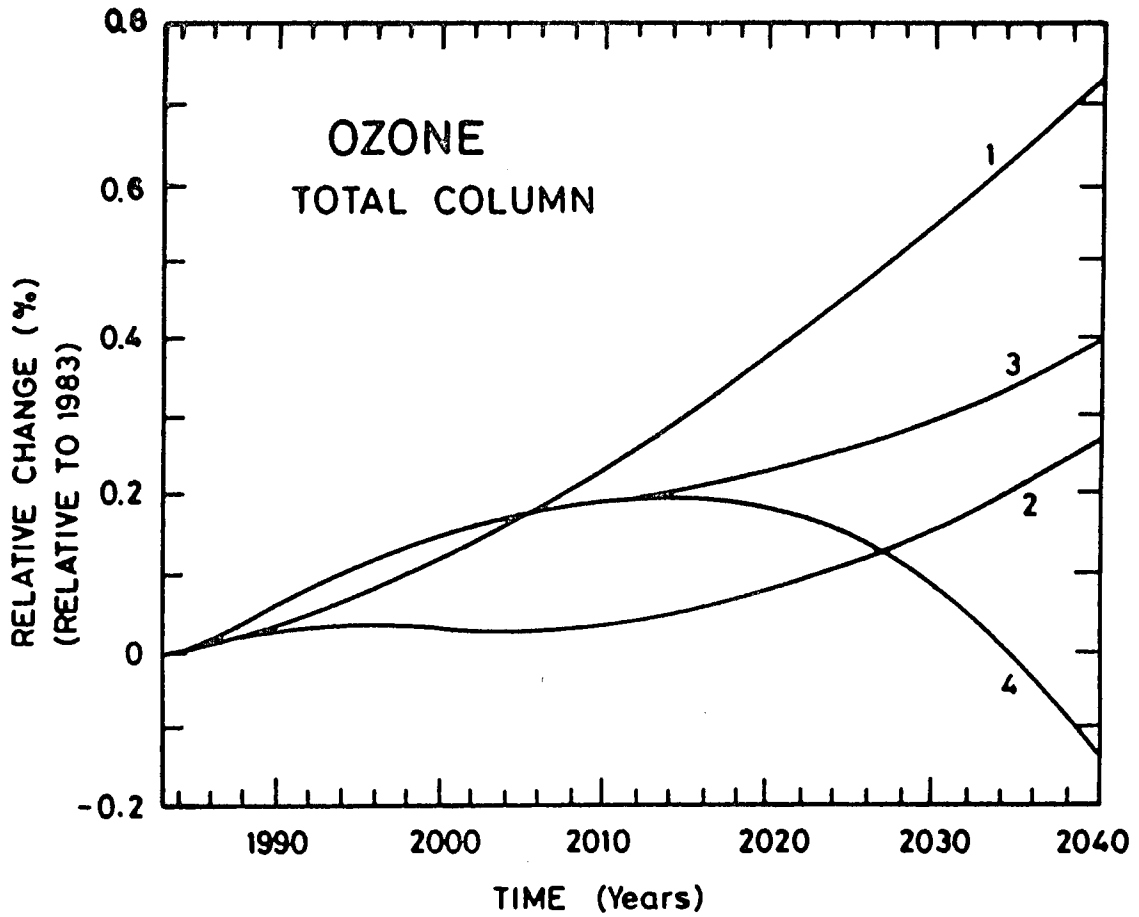lative values.

Figure 23 Future relative change in the total ozone amount as a function of time, computed in the 4 different cases.

the cases where all gases and only CFCs are considered is depicted in figure 24. The effect of a large aircraft emission can be estimated from figures 25 (scenario 1) and 26 (scenario 2).
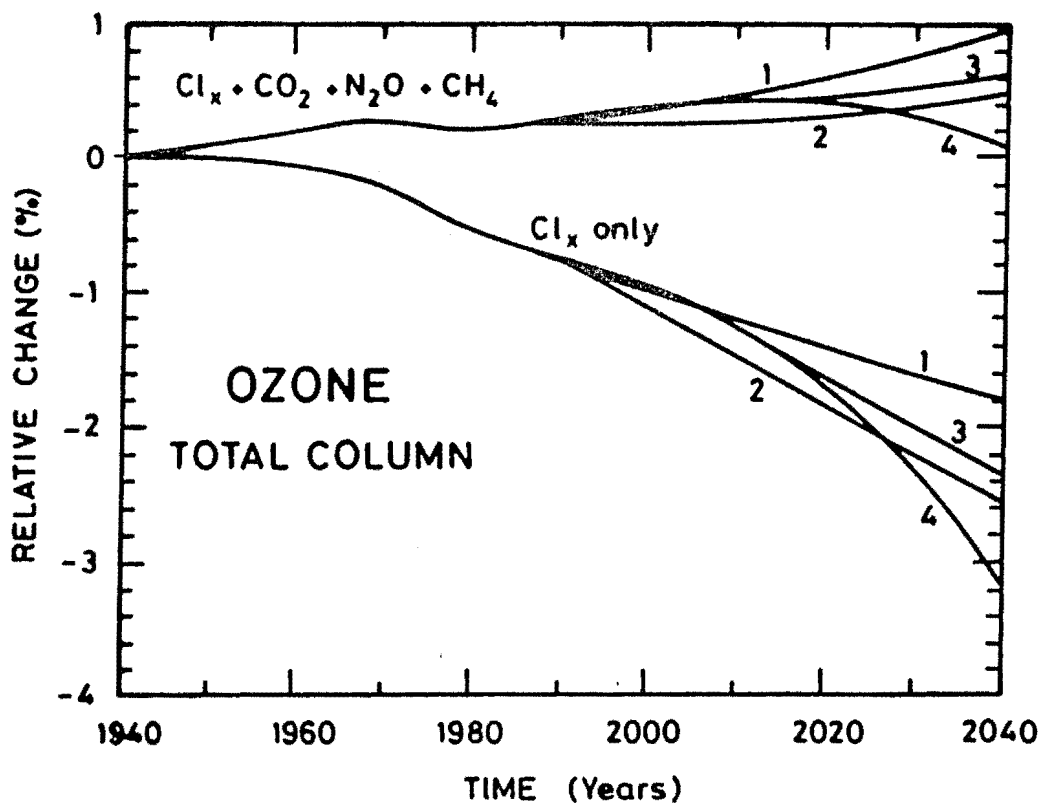
<u>Figure 24</u> Relative change in the total ozone concentration, as a
function of time from 1940, computed in the 4 different
scenarios and compared to the results of a similar calculation
considering only the corresponding perturbations due to the
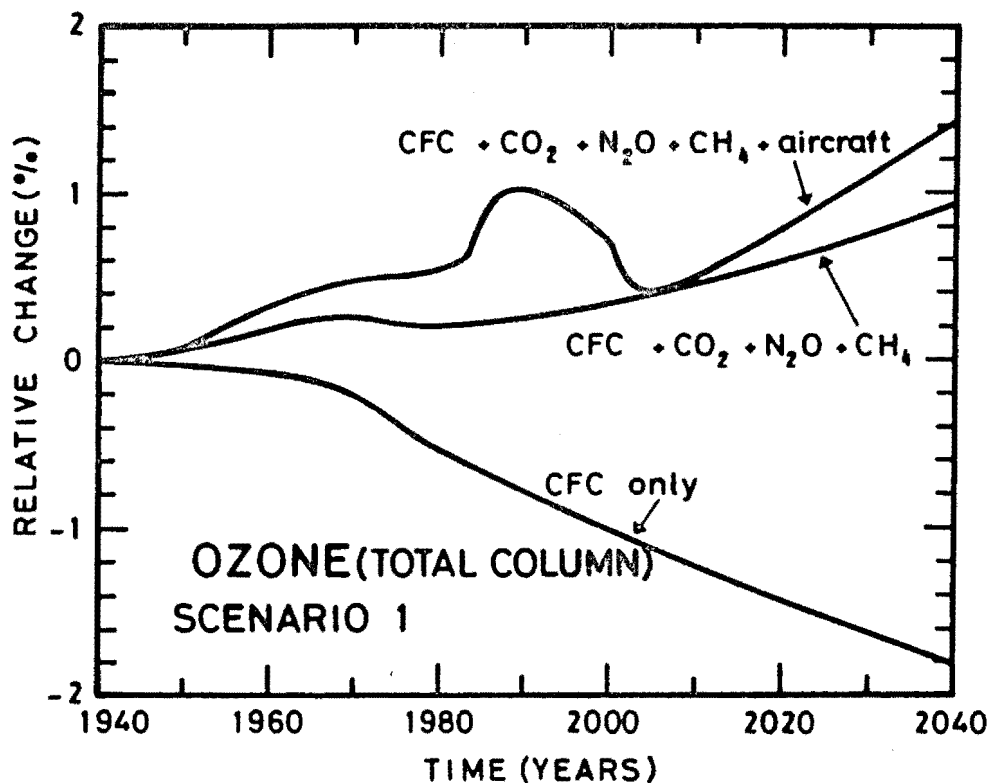emission of chlorocarbons.

Figure 25 Relative change in the integrated ozone concentration, as a
function of time from 1940 to 2040, obtained for scenario 1
and compared to (1) a case considering only the same chloro-
carbons perturbation and to (2) a case where an injection of
$NO_x$ by aircraft engines is assumed in addition to the other
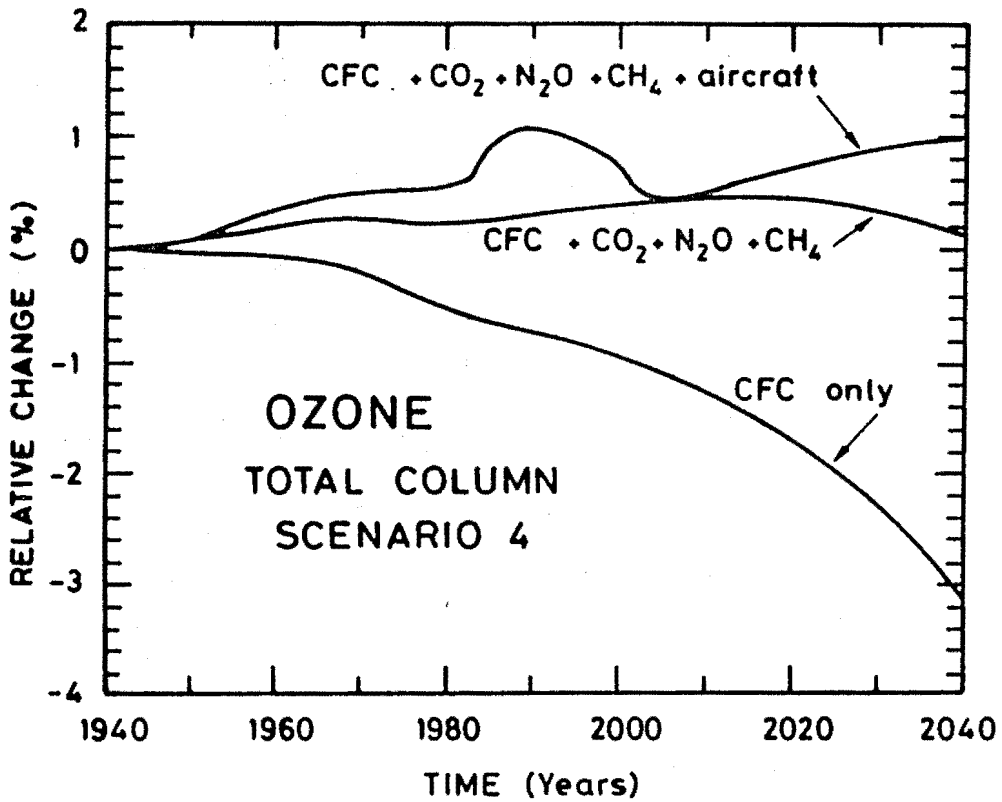perturbations included in the scenario.

**Figure 26** Same as figure 25 but for scenario 4.