

I N S T I T U T D ' A E R O N O M I E S P A T I A L E D E B E L G I O U E

3 - Avenue Circulaire

B - 1180 BRUXELLES

# **AERONOMICA ACTA**

A - N° 288 - 1984

**Ozone and temperature trends due to the injection of  
trace species in the atmosphere**

by

**Guy BRASSEUR**

B E L G I S C H I N S T I T U U T V O O R R U I M T E - A E R O N O M I E

3 - Ringlaan

B - 1180 BRUSSEL

## FOREWORD

This paper has been presented at a seminar organized by the Commission of the European Communities (Climate Program). It will be published in the proceedings of the meeting.

## AVANT-PROPOS

Cette note a été présentée à un séminaire organisé le 18 mai 1984 par la Commission des Communautés Européennes (Programme Climatique). Elle sera publiée dans les compte-rendus de la réunion.

## VOORWOORD

Deze tekst werd voorgesteld tijdens een werkvergadering die op 18 mei 1984 georganiseerd werd door de Commissie van de Europese Gemeenschappen (Klimaatprogramma). Hij zal gepubliceerd worden in de verslagen van de bijeenkomst.

## VORWORT

Dieser Text wurde präsentiert während der Tagung organisiert am 18 Mai 1984 von der Kommission der Europäische Gemeinschaften (Klimaprogramm). Er wird publiziert werden in die Rapporten der Tagung.

OZONE AND TEMPERATURE TRENDS DUE TO THE INJECTION OF  
TRACE SPECIES IN THE ATMOSPHERE

by

Guy BRASSEUR

Abstract

The paper presents a model of the possible future ozone and temperature changes in relation with the injection in the atmosphere of several trace species produced by man-made activity.

Résumé

Cette note présente les résultats d'un modèle qui prédit les changements de la quantité d'ozone et de la température sous l'effet de l'injection dans l'atmosphère de gaz produits par l'activité humaine.

## Samenvatting

Deze tekst geeft de resultaten van een model dat de veranderingen in de temperatuur alsook in de hoeveelheid ozon voorziet onder invloed van de injectie in de atmosfeer van verscheidene gassen die door menselijke activiteiten geproduceerd worden.

## Zusammenfassung

Dieser Text gibt die Resultaten eines Modelles das die Veränderungen in der Temperatur wie auch in der Quantität Ozon voraussetzt unter dem Einfluss von der Injektion in der Atmosphäre von verschiedenen Gasen produziert von menschlichen Aktivitäten.

## I. INTRODUCTION

Much attention is presently given to the potential effects of gases which are released by the industry and penetrate in the stratosphere where they interact with ozone. Among these gases, chlorofluorocarbons (CFC), methane ( $\text{CH}_4$ ), nitrous oxide ( $\text{N}_2\text{O}$ ) and carbon dioxide ( $\text{CO}_2$ ) play an important role. Their radiative and chemical effects are currently studied by means of coupled chemical/radiative models (Wuebbles, 1983; Brasseur and De Rudder, 1984; Brühl and Crutzen, 1984, etc...) which take into account a large number of chemical and photochemical reactions and consider the effect of optically active molecules on emission and absorption processes.

The purpose of this study is to present and discuss data obtained with such a model, considering plausible scenarios for the emission of the optically and chemically active constituents. The time dependent simulation refers to a preindustrial atmosphere (year 1850) and predicts the effects of these gases until year 2080.

The model described elsewhere (Brasseur et al, 1982; Brasseur and De Rudder, 1984) includes a detailed chemical and photochemical scheme, taking into account the action of oxygen, hydrogen, nitrogen, chlorine and carbon species. The vertical temperature profile is derived from a thermal scheme in which the heating is calculated using the parameterization suggested by Schoeberl and Strobel (1978). The radiative transfer in the infrared is treated by determining the transmission function in 4 large spectral intervals. The upward and downward infrared flux as well as the corresponding cooling rates are obtained by solving the radiative transfer equation in which the transmission functions are parameterized as a function of the amount of absorbing gases. This parameterization is based on a study made with a multispectral radiative model by Morcrette (private communication). An energy balance is achieved at the top of the atmosphere assuming a global earth-atmosphere albedo of 0.3. The model extends from 0 to

70 km. The vertical transport of the trace species and of the heat (especially in the troposphere where convective instability appears) is parametrized by means of an eddy diffusion coefficient.

## II. ADOPTED SCENARIO FOR THE MODEL CALCULATION

A prognostic of the future state of the atmosphere requires the introduction in the model of a realistic scenario. Because of the large uncertainties in the projected emissions of the pollutants, the following scenario should be considered as an example based on realistic considerations. Figure 1 shows the historical data and the projected values which have been used for the industrial sources of the several halocarbons which are released in the atmosphere (Logan *et al.*, 1978). The corresponding CIX mixing ratio at the stratopause from year 1950 to year 2080 is reproduced in figure 2. It reaches 9 ppbv in year 2080, that is a factor of 3 larger than the present value.

The CO<sub>2</sub> amount in the atmosphere during the next century is not straightforward to predict but a number of scenarios for the related emission, based on the projected fossil fuel usage have been developed (Niehaus, 1976; Rotty, 1977 etc...). We have assumed for the pre-industrial atmosphere (1850) a mixing ratio of 270 ppmv and, after year 1979 a growth given by (Wuebbles *et al.*, 1983)

$$f(\text{CO}_2) = 335.0 \exp [0.0056 (t - 1979)]$$

where  $f(\text{CO}_2)$  is the CO<sub>2</sub> mixing ratio expressed in ppmv and  $t$  the year under consideration.

Atmospheric measurements carried out in the last decade have shown a systematic increase in the N<sub>2</sub>O concentration of about 0.25 percent per year (see e.g. Weiss, 1981; Khalil and Rasmussen, 1981;

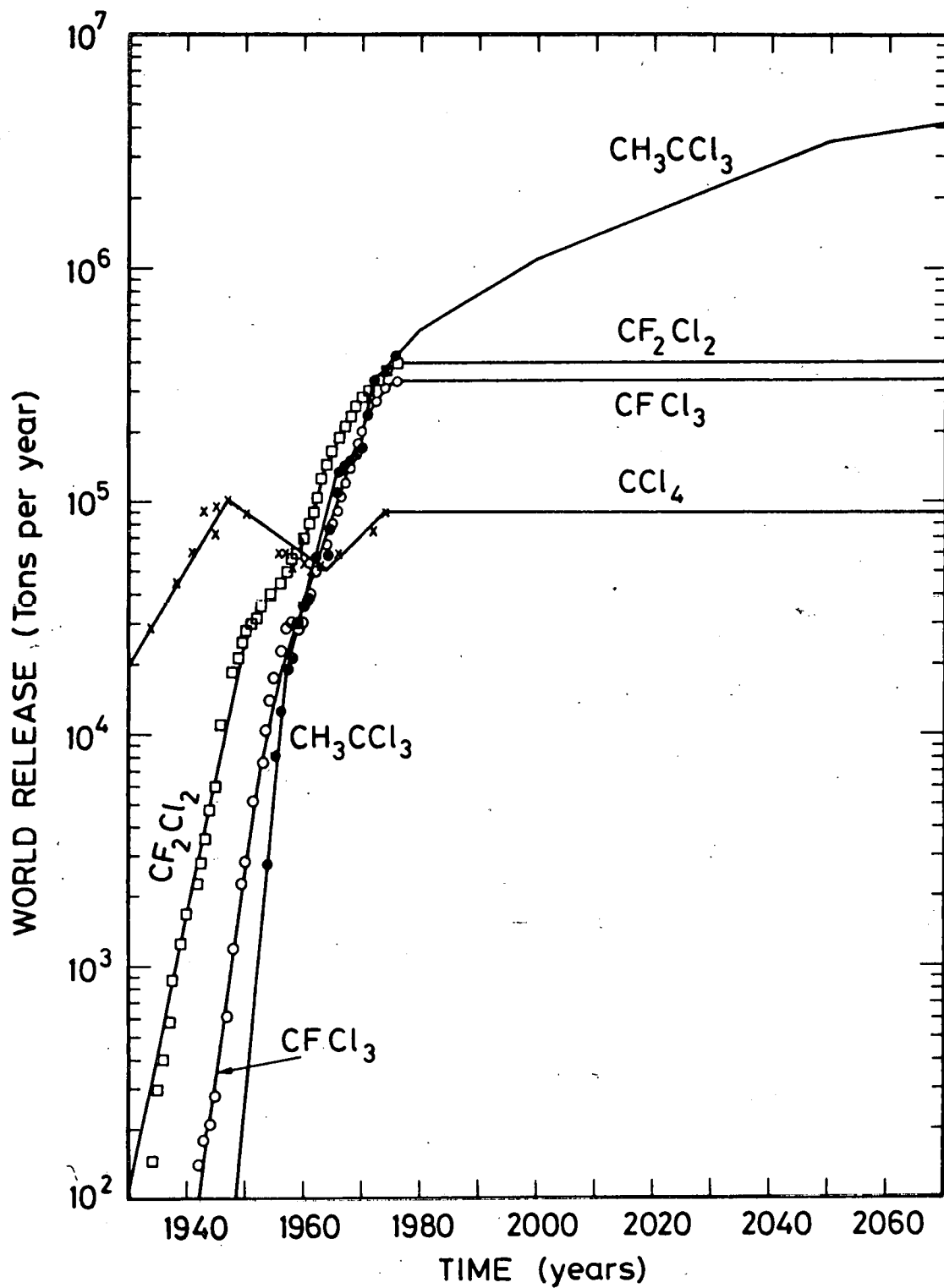


Fig. 1.- Historical release and assumed future emission of different halocarbons adopted as a scenario for the perturbation calculations, after Logan et al., 1978.

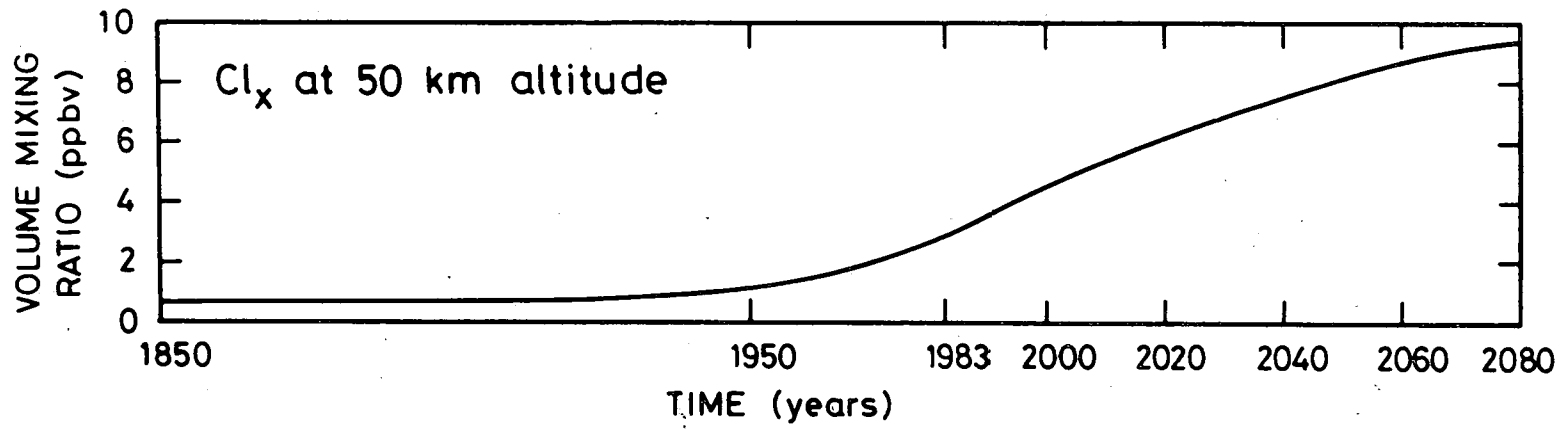


Fig. 2.- Variation of the Cl<sub>x</sub> mixing ratio in the upper stratosphere induced by the halocarbons release adopted in the scenario and described above (see fig. 1).



1983). Such a growth has been assumed to remain constant in the future until year 2080. The N<sub>2</sub>O mixing ratio in year 1850 is assumed to be equal to 285 ppbv. The value in 2080 is 425 ppbv.

The existence of a systematic trend in the methane amount is subject to discussion. Analyzing a series of measurements made in the past decade and taking into account several corrections made for older data, Ehhalt (1983) does not derive any significant change in the CH<sub>4</sub> mixing ratio. However, continuous observations made in the recent years by Rasmussen and Khalil (1981), Blake et al. (1982) and Rowland (1983) show an increase of 1 to 2 percent per year. A value of 1 ppmv has been adopted for the preindustrial mixing ratio with a increase of the order of 1.5 percent/yr for the future, leading to a mixing ratio of 7.5 ppmv in year 2080. The perturbations of CH<sub>4</sub>, N<sub>2</sub>O and CO<sub>2</sub> used as input for the model are given as a function of time in figure 3.

Aircraft emissions have been introduced after year 1950 and are similar to that introduced by Wuebbles et al. (1983). These numbers (see fig. 4) are based on the analysis by Bauer, 1978 and Oliver et al., 1977. The corresponding trend between 1950 and 2000 may be somewhat overestimated due to the flight reductions in relation with the economic crisis and the increase in the fuel costs. This particular scenario should therefore correspond to an upper limit in the possible ozone change. After year 2000, the aircraft emission is supposed to remain constant. The key parameter in this type of calculation is the ratio between artificial and natural NO<sub>x</sub> sources. The value of this parameter as a function of time is highly uncertain. The adopted sources may lead to an overestimation of the ozone increase in the troposphere.

### III. RESULTS AND DISCUSSION

The changes which are calculated and discussed below use, as a reference, the preindustrial atmosphere of year 1850. The variations

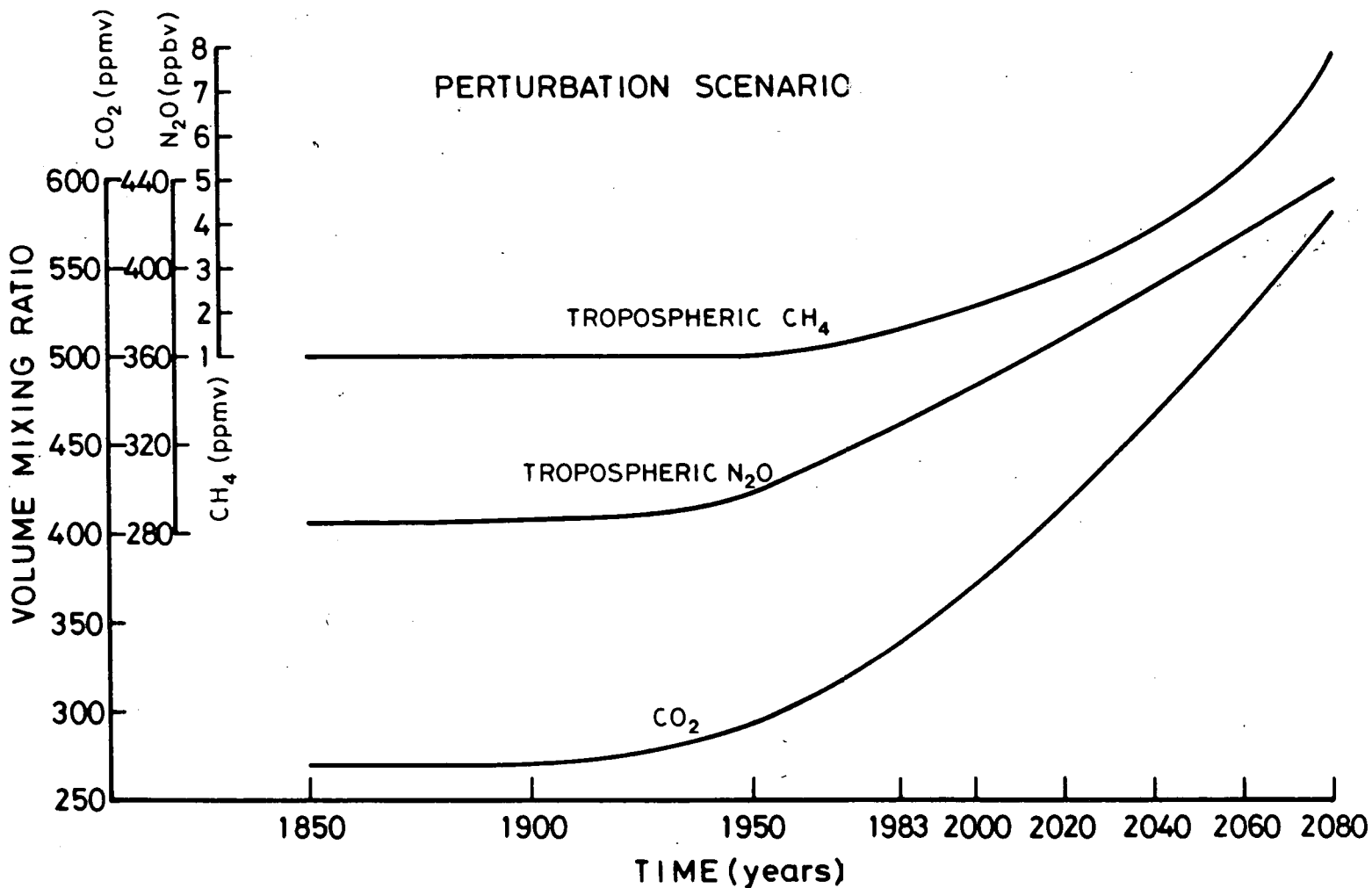


Fig. 3.- Assumed evolution of the N<sub>2</sub>O, CO<sub>2</sub> and CH<sub>4</sub> mixing ratio from the pre-industrial era to year 2080. These curves correspond to an increase in N<sub>2</sub>O of 0.3% per year from the present day, of 0.56%/yr for CO<sub>2</sub> and of 1.5%/yr for CH<sub>4</sub>.

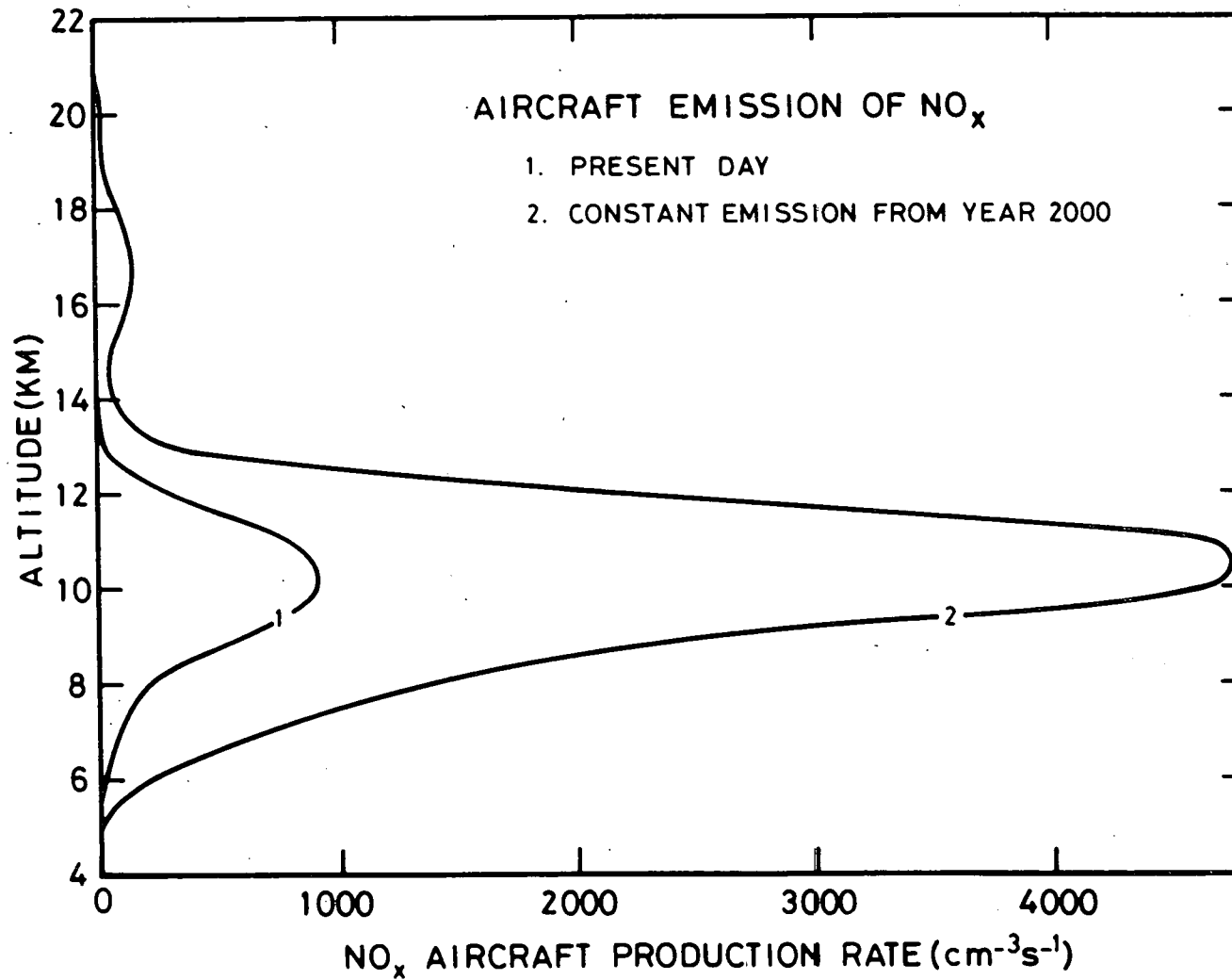


Fig. 4.- Assumed present and future release due to aircraft operations. Curve 1 corresponds to the present day situation and is derived from the analysis by Bauer (1978). The projected emission for year 2000 shown by curve 2 is similar to the emission suggested by Oliver et al. (1977) for year 1990. A linear trend is assumed before 2000. After this period, the NO<sub>x</sub> release remains constant.

in total ozone and temperature at the Earth's surface for the scenario described above are listed in the table 1.

This table clearly indicates that the surface temperature increases regularly due essentially to the enhancement of CO<sub>2</sub> (greenhouse effect). This temperature change is probably somewhat underestimated, at least for the future conditions, since the direct radiative effect of N<sub>2</sub>O, CH<sub>4</sub> and CFCs is not yet included in the model. It is generally assumed (WMO, 1983) that the effect of a doubling of these gases could be as large as the contribution of a doubling in CO<sub>2</sub>. The model also indicates that the present temperature should be 0.7 K above its preindustrial value. Such a number is difficult to compare with observed data since the "random" perturbations due to volcanic activity which probably lead to a cooling of the Earth's surface (see Brasseur and Solomon, 1984 for a discussion) are not included in the model although the corresponding effects might be larger than the present CO<sub>2</sub> effects.

Total ozone, as shown in table 1, varies by a few percent only and the depletion is never larger than 1 percent. In fact, as indicated by figures 5a and b, the small change in the column results from the difference between a significant ozone depletion in the stratosphere and a rather similar increase in the troposphere. An examination of fig. 5a shows that ozone is rather stable in the 20-30 km region. The largest relative depletion at 40 km is due to the action of CFCs. The resulting ozone reduction in the upper stratosphere increases with time until year 2020. After this time, the enhancement in the methane concentration reduces the amount of the active Cl<sub>x</sub> (Cl and ClO) in favor of the inactive reservoir HCl by reaction

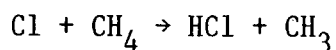


TABLE 1.- Changes in the ozone column  $\Delta O_3/O_3$  and in the temperature at the Earth's surface ( $\Delta T_s$ ) as a function of time.

Year	$\Delta O_3/O_3$ (percent)	$\Delta T_s$ (K)
1950	+ 0.5	+ 0.2
1983	+ 0.1	+ 0.7
1990	- 1.0	+ 0.8
2000	- 1.0	+ 1.0
2020	- 0.6	+ 1.3
2040	0.0	+ 1.6
2060	+ 1.0	+ 2.0
2080	+ 3.0	+ 2.3

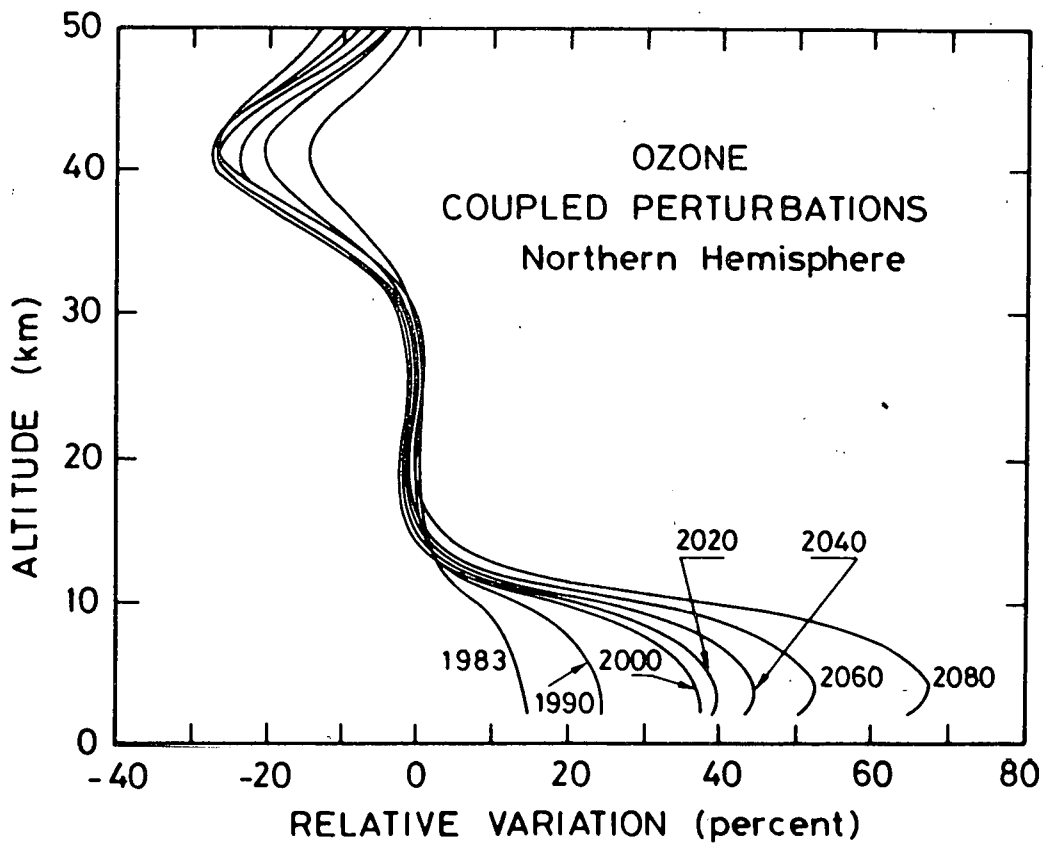


Fig. 5a.- Relative variation in the ozone concentration with regard to its preindustrial value, corresponding to the adopted perturbations scenario.

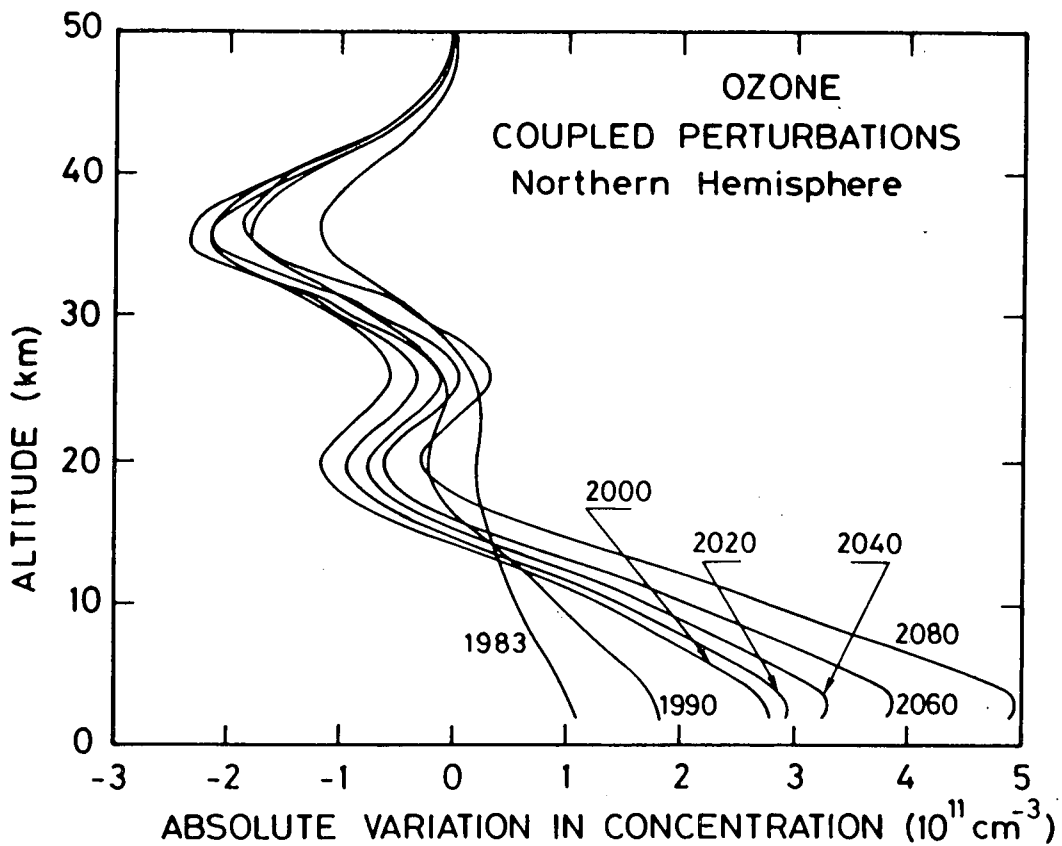


Fig. 5b.- Calculated deviation of the ozone concentration from its preindustrial value, corresponding to the adopted perturbations of fig. 1-4.

The ratio ClO/HCl decreases and the efficiency of the destruction of ozone by  $Cl_x$  is considerably reduced. As discussed below, the simultaneous increase in carbon dioxide also lessens the ozone depletion.

The reduction of  $O_3$  in the lower stratosphere (relatively small but rather significant in absolute value) is due to the increase in the  $N_2O$  and in the associated  $NO_x$  amount. This perturbation is small due to the relatively slow prescribed increase in the  $N_2O$  concentration. Moreover, the dramatic enhancement in methane leads to a larger OH concentration and consequently a lower  $NO_2/HNO_3$  ratio. This effect stabilizes ozone in the middle stratosphere.

The rather fast increase in tropospheric ozone which almost counterbalances the decrease in the stratosphere is due, for the period prior to 2000, to the rapidly varying aircraft emission. After year 2000 the  $NO_x$  injection is assumed to remain constant and the slower increase has to be attributed to the enhancement in the methane concentration.

Sensitivity calculations show that the magnitude of the methane effect on ozone depends much on the amount of  $NO_x$  present in the troposphere. It is therefore required to better understand the absolute values of the tropospheric production rates of  $NO_x$  by lightning, decomposition of PAN, oxidation of  $NH_3$  or any other process. In the present model study, we have adopted a constant production of  $1000 \text{ cm}^{-3} \text{ s}^{-1}$  below 10 km, corresponding to a world production of  $10^6$  Tons N/yr.

Carbon dioxide has no direct chemical effect below 70-80 km. It has however a radiative influence and its predicted upward trend should progressively cool the upper stratosphere. Because of the chemical/thermal interaction, a given decrease in the stratopause temperature should increase the local ozone concentration. Consequently, the adopted scenario for the  $CO_2$  evolution should stabilize ozone against the CFC perturbation.

The preceding results refer to simultaneous effects of various emissions. The response of the atmosphere to individual perturbations has also been considered. When, for example, only a continuous injection of CFCs is considered (fig. 1), the ozone amount decreases gradually and the corresponding depletion reaches 3 percent in year 2080. This value does not yet correspond to a steady state. A doubling of nitrous oxide reduces the ozone column by 8 percent and a doubling of methane by about 2 percent. When the amount of carbon dioxide is doubled, the ozone column is increased by 3 percent and the local  $O_3$  concentration at 40 km is enhanced by about 23 percent.

The temperature in the stratosphere is very sensitive to the chemical composition and especially to the vertical distribution of ozone and carbon dioxide. Both the reduction of  $O_3$  (due essentially to large  $Cl_x$  amounts) and the enhancement of  $CO_2$  lead to a decrease in the temperature. Contrary to what happens for the ozone concentration, the simultaneous introduction of the two perturbations does not stabilize the temperature but leads to a larger cooling. As shown by figure 6, a reduction of more than 20 K should be expected at the stratopause. The modification in the vertical temperature gradient will infer changes in the general circulation patterns. These dynamical effects require, for their study, multidimensional investigations.

## CONCLUSIONS

Model calculations show that the injection of several gases released by man-made activity could significantly modify the ozone concentration and the temperature in the stratosphere. These changes should therefore lead to modifications in the strength of the atmospheric dynamics and in the surface temperature. The problem discussed in this paper, because of its climatic implications, requires much attention from the scientific community as well as from the decision makers.



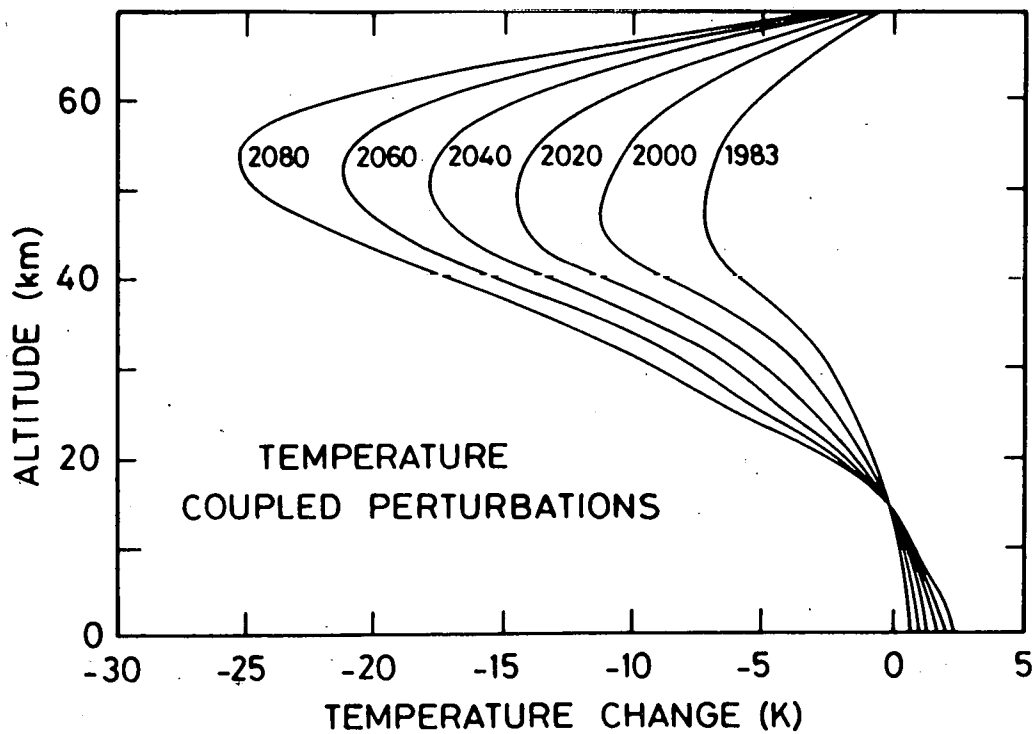


Fig. 6.- Calculated change in the temperature from the preindustrial era, for several years running from the present day to year 2080, corresponding to the scenario described in figures 1-4.

It should be emphasized that the variations calculated in the model discussed above results from a number of non-linear interactive processes which are not yet completely understood. For example, numerical simulations (Prather et al, 1984) indicate that above a certain amount of  $Cl_x$ , the sensitivity of ozone to odd chlorine increases suddenly, leading rapidly to a large ozone depletions. This non-linear effect between  $O_3$  and  $Cl_x$  is controled by odd nitrogen : at high chlorine, the removal of ozone is dominated by reactions involving  $ClO$  while, at low chlorine, it is regulated by reactions with  $NO_2$ . Detailed analyses are required to study such coupled processes with their implications on the future climate.

#### ACKNOWLEDGEMENTS

I wish to thank A. De Rudder and Chr. Tricot for several profitable discussions and help in the computation work. This work was supported by the Chemical Manufacturers Association (CMA).

## REFERENCES

- BAUER, E., A catalogue of perturbing influences on stratospheric ozone, 1955-1975, US-FAA Report N° FAA-EQ-78, 20, US Dept. of Transportation, Washington DC, September 1978.
- BLAKE, D.R., E.W. MAYER, S.C. TYLER, Y. MAKIDA, D.C MONTAGNE and F.S. ROWLAND, Global increase in atmospheric methane concentrations between 1978 and 1980, *Geophys. Res. Lett.*, 9, 477, 1982.
- BRASSEUR, G., A. DE RUDDER and A. ROUCOUR, The natural and perturbed ozonosphere, Proceedings of the International Conference on Environmental Pollution, Thessaloniki, 840, 1982.
- BRASSEUR, G. and A. DE RUDDER, Agents and effects of ozone trends in the atmosphere, Proceedings of the NATO workshop on "The impact of solar ultraviolet (UV-B) radiation upon terrestrial ecosystems. I. Agricultural systems, Bad Windsheim, FRG, September 27-30, 1983; Springer Verlag, to be published, 1984.
- BRASSEUR, G. and S. SOLOMON, *Aeronomy of the middle atmosphere*, Reidel Publishing Co, Dordrecht, The Netherlands, 441 pp, 1984.
- BRUHL, C. and P.J. CRUTZEN, Radiative-convective model to study the sensitivity of climate and chemical composition to a variety of human activities, this issue, 1984.
- EHHALT, D.H., R.J. ZANDER and R.A. LAMONTAGNE, On the temporal increase of tropospheric CH<sub>4</sub>, *J. Geophys. Res.*, 88, 8442, 1983.
- KHALIL, M.A.K. and R.A. RASMUSSEN, Increases in atmospheric concentrations of halocarbons and N<sub>2</sub>O, *Geophys. Monit. Clim. Change*, 9, 134, 1981.
- KHALIL, M.A.K. and R.A. RASMUSSEN, Increase and seasonal cycles of nitrous oxide in the Earth's atmosphere, *Tellus*, 35, 161, 1983.
- LOGAN, J.A., M.J. PRATHER, S.C. WOFYSY and M.B. McELROY, *Atmospheric chemistry : response to human influence*, *Phil. Trans. Roy. Soc. Lond.*, 290, 187, 1978.

- NIEHAUS, F., A non-linear eight level random model to calculate future CO<sub>2</sub> and C-14 burden to the atmosphere, Int. Inst. for Applied Systems Analysis Research Memorandum, RM 76-35, 1976.
- OLIVER, R.C., E. BAUER, H. HIDALGO, K.A. GARDNER and W. WASYLIKISWSKYJ, Aircraft emissions : Potential effects on ozone and climate, US Dept. of Transportation Report FAA-EQ-77-3, 1977.
- PRATHER, M.J., M.B. McELROY and S.C. WOFSY, Reductions in ozone at high concentrations of stratospheric halogens, Paper presented at the WMO/NASA Workshop, Starnberger See, FRG, June 11-16, 1984.
- RASMUSSEN, R.A. and M.A.K. KHALIL, Atmospheric methane (CH<sub>4</sub>) Trends and seasonal cycles, J. Geophys. Res., 86, 9826, 1981.
- ROWLAND, F.S., private communication, 1983.
- ROTTY, R.M., The atmospheric CO<sub>2</sub> consequences of heavy dependence on coal, Oak Ridge National Laboratory Report, ORAU/IEA-77-27, 1977.
- SCHOEBERL, R.M. and D.F. STROBEL, The zonally average circulation of the middle atmosphere, J. Atm. Sci., 35, 577, 1978.
- WEISS, R.W., The temporal and spatial distribution of tropospheric nitrous oxide, J. Geophys. Res., 86, 7185, 1981.
- WUEBBLES, D.J., F.M. LUTHER and J.E. PENNER, Effect of coupled anthropogenic perturbations on stratospheric ozone, J. Geophys. Res., 88, 1449, 1983.