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Critical Analysis of Recent Reports on the Effect of Chlorofluorocarbons on Atmospheric Ozone

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FOREWORD

According to American and European scientists, chlorofluorocarbons released during their use as aerosol propellants, refrigerants and foam-blowing agents migrate slowly to the stratosphere where they could destroy the ozone which is protective barrier agains UV-B radiation.

This problem cannot be ignored and has become an increasing concern for the responsible authorities. Both on a national and international scale every effort is being made to obtain the knowledge needed for a sensible evaluation of the impact of chlorofluorocarbons on atmospheric ozone.

On 26 March 1980 the Council of Ministers of the European Communities adopted a decision concerning chlorofluorocarbons in the environment[1]. The first part of the decision laid down that industry situated on the territory of the Member States should not increase its production capacity for F-11 and F-12 and must reduce, by 31 December 1981 the amount of chlorofluorocarbons used in aerosols by at least 30% of 1976 levels.

Article 2 of the Decision required that in the course of the first half of 1980, the measures taken had to be re-examined in the light of the scientific and economic data available. In view of this re-examination, the Commission asked Dr. Guy BRASSEUR to prepare a report comparing and analysing recent studies on the effect of chlorofluorocarbons on atmospheric ozone.

Amongst others, two important studies on this matter were published in 1979; one was published by the United Kingdom Department of the Environment and the other by the United States National Academy of Sciences. The ozone depletion figures obtained in the United Kingdom and

in the United States are comparable. However, the difference between the two approaches lies in the confidence placed in the models and the degree of simplification considered acceptable. It is of the first importance to be able to understand these differences within the scientific community.

Dr. Brasseur discusses in his report the different points of view contained in the various studies as well as the weights given to the uncertainties in the various models. He gives us a critical insight into the current state of scientific knowledge of ozone depletion rather than new findings or a new assessment of the problem. Dr. Brasseur concludes his report by noting what steps should be taken in the near future to continue research in this area.

His report was submitted to the Council of Ministers of the Environment of the European Communities in June 1980(2).

M. Carpentier
Director-General
Environment and Consumer
Protection Service

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(1) Council Decision 80/372/CEE, O.J. L 90 of 3.4.80

(2) Communication from the Commission to the Council, Chlorofluorocarbons in the Environment, COM (80) 339.

This report is an english translation of the original french version.

SUMMARY

CRITICAL ANALYSIS OF RECENT REPORTS ON THE EFFECT OF CHLOROFLUOROCARBONS ON ATMOSPHERIC OZONE

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1. INTRODUCTION

During 1979, a number of reports on the likely effect of chlorofluorocarbons on atmospheric ozone were prepared. One of these, entitled "Chlorofluorocarbons and their effect on stratospheric ozone" was published in the autumn of 1979 by the United Kingdom Department of the Environment (Central Directorate on Environmental Pollution). It is in two parts :

I. A report by the Directorate for Air, Noise and Wastes of the Department of the Environment,

II. A report by the Stratospheric Research Advisory Committee (a scientific committee chaired by Dr. Murgatroyd).

Of particular interest is the second report which presents a consensus view of the British scientific community specializing in atmospheric physics. It will be referred to subsequently as the "UK report".

A second report was drawn up by a panel on "Stratospheric Chemistry and Transport" chaired by Dr. Schiff and published in November 1979 by the United States Academy of Sciences. It will be referred to as the "NAS report" (National Academy of Sciences).

A more comprehensive study covering not only the problem of the stability of the ozonosphere but also the effects of ozone depletion (biological effects, effects on health, climatic effects) and the possible alternatives to chlorofluorocarbons was published at the end of 1979 by the United States Academy of Sciences : the committees responsible for the report were : the "Committee on Impacts of stratospheric Changes" (Chairman : Dr. J.W. Tukey) and the "Committee on Alternatives for the Reduction of Chlorofluorocarbon Emissions" (Chairman : Dr. M.S. Peters). As to the causes of potential ozone depletion, this document reiterates the conclusions of the NAS report (Dr. Schiff) to which we have referred. The problem of the multiple effects of a reduction in the amount of ozone will not be dealt with here since it is outside our terms of reference.

Another very interesting report was published by NASA (Hudson and Reed) on 1 January 1980. It is the outcome of a joint project undertaken by some hundred scientists in the United States. It will be known as the "NASA report".

In December 1979, the Federal Aviation Agency (FAA) published a report under its "High Altitude Pollution Program" (HAPP) on the protection of ozone as provided for in the "Clean Air Act Amendments" of 1977. This study was carried out by a committee chaired by Professor F.S. Rowland. The European Communities (DG 12) are preparing reports on the work being done in Europe on mathematical models of the atmosphere and on laboratory measurements of the parameters essential to the study of atmospheric chemistry.

It should be pointed out that other papers have been published in recent years but, in a number of cases, the findings are now out of date. For information purposes, however, Table 1 contains a bibliography of official reports on the effect of chlorofluorocarbons (CFC) on atmospheric ozone. These documents contain references to the relevant articles published in the scientific literature. Under the auspices

of the United Nations Environment Programme (UNEP) a key meeting of the Coordinating Committee on the ozone layer was held in Paris from 20 to 23 November 1979. The governments represented each reported on the work being carried out in their countries. A number of conclusions were recached and recommendations put forward.

The main findings of the various reports and of the UNEP are contained in the Appendices. This will provide the reader with a rapid summary of the most recent studies.

TABLE 1.- Official reports and summary records of international conferences on the problem of chlorofluorocarbons.

Department of the Environment, Central Unit on Environmental Pollution, Chlorocluorocarbons and their effect on stratospheric Ozone, Pollution Paper No 5, London, 1976.

National Academy of Sciences, Halocarbons : Environmental Effects of Chlorofluoromethane Release, Committee on Impacts of Stratospheric Change, Assembly of Mathematical and Physical Sciences, National Research Council, Washington, D.C. 1976.

National Aeronautics and Space Administration, Chlorofluoromethanes and the Stratosphere, Robert D. HUDSON, Goddard Space Flight Center, Greenbelt, Maryland, 1977.

National Academy of Sciences, Response to the Ozone Protection Sections of the Clean Air Act Amendments of 1977 : An Interim Report, Committee on the Impacts of Stratospheric Change, Assembly of Mathematical and Physical Sciences, National Research Council, Washington, D.C., 1977.

Department of the Environment, Central Directorate on Environmental Pollution, Chlorofluorocarbons and their effect on stratospheric ozone, (Second Report) Part 1 : Report by the Directorate of Air, Noise and Wastes, Department of the Environment; Part 2 : Report to the Department of the Environment by the Stratospheric Research Advisory Committee, London 1979.

National Academy of Sciences, Stratospheric Ozone Depletion by Halocarbons : Chemistry and Transport, Panel on Stratospheric Chemistry and Transport, Committee on Impacts of Stratospheric Change, Assembly of Mathematical and Physical Sciences, National Research Council, Washington, D.C., 1979.

National Academy of Sciences, Protection against Depletion of Stratospheric Ozone by Chlorofluorocarbons, Committee on Impacts of Stratospheric Change, Assembly of Mathematical and Physical Sciences, Committee on Alternatives for the Reduction of Chlorofluorocarbon Emissions, Commission on Sociotechnical Systems, Washington, D.C., 1979.

Federal Aviation Administration, High Altitude Pollution Program, Second Biennial Report prepared in accordance with the ozone protection provision, Section 153 (g), of the clean air act amendments of 1977, Washington, D.C. 1979.

National Aeronautics and Space Administration, The Stratosphere : Present and Future, Robert D. HUDSON and Edith I. REED, NASA Goddard Space Flight Center, 1979.

Commission of the European Communities, Directorate-General for Research, Science and Education, Environmental Research Programme 1976-1980, Research on the ozone shield depletion problem under CEC coordination, Proceedings of the Second Meeting of the Study Groups, Brussels 1978.

Internationale Konferenz über Fluorchlorkohlenwasserstoffe, Konferenz-ergebnisse, Vol. 1, Umweltbundesamt, München, 1978.

Internationale Konferenz über Fluorchlorkohlenwasserstoffe, Konferenz-ergebnisse, Vol. 2, Umweltbundesamt, München, 1978.

It should be pointed out that the three main American reports referred to (NAS, NASA, FAA) were conceived, prepared and drafted by committees composed - in part at least - of the same scientists. Although the opinions given sharply differ in certain cases, the underlying philosophy is the same. It reflects a consensus view of the American scientific community. The UK report, on the other hand, drafted by researchers in the United Kingdom has an original approach compared with that of the Americans. The research was carried out independently, the conclusions occasionally differ, and the analysis of the findings leads to reservations about the American position.

The aim of this paper is to attempt to analyse the conclusions reached by the various reports in order to take stock of the certainties and uncertainties surrounding the problem of ozone. An attempt will then be made to put forward a number of projects to be carried out within the next five years.

2. MAIN CONCLUSIONS OF THE RESEARCH INTO THE EFFECT OF CFCs ON OZONE

2.1. US findings

The findings obtained by the researchers working on the NASA project may be summarized very briefly by saying that the most probable depletion in the total quantity of ozone, assuming continued emission of CFC 11 and CFC 12 at 1975 level, would be between 15 and 18% when the equilibrium state is reached. According to NASA, the depletion will lie

in the range 7.5 to 29.8% - with a probability of 95% - given the uncertainties concerning the chemical and photochemical parameters.

The NAS Report concludes that the most likely depletion will be 16.5%. This figure is based on a value of 18.6% given by the mathematical models and takes account of additional effects arising principally from the possible destruction of CFCs in the troposphere and the greenhouse effect of these industrial compounds. The uncertainties affecting the parameters used in the calculations led the NAS to estimate the ozone depletion as lying in the range 5 to 28% with a 95% confidence level.

There is thus a broad measure of agreement between the NAS and NASA findings : they both point to an adverse effect of CFCs on the ozone layer and the average figure assigned to the depletion may be put at 16.5% ± 10%. The statistical error is thus quite high.

2.2. UK findings

The UK Report challenges the assumption of assigning a global (or average) figure to atmospheric ozone depletion in view of the complex nature of the atmosphere and the existence of many natural variables. The likelyhood therefore is a relative reduction of O_3 which varies with latitude and season. But if the calculation method used by the American Researchers is adopted, the following values are obtained : 13% in the Harwell model, 11% in the Meteorological Office model (2-D) and 16% in the Oxford model (2-D).

To appreciate the difficulties inherent in these models, however, it will be observed that by varying the profile of the vertical transport coefficient (at present poorly defined), the result given by the Harwell model may differ by between 6 and 22%. The two-dimensional models also display a high degree of sensitivity to the values assigned to the transport parameters.

Lastly, the UK Report lays emphasis on the likely interaction with other human activities (emission of CO_2 from combustion processes or N_2O from chemical fertilizers). It would seem that these effects appreciably alter the calculations of ozone depletion by CFCs.

2.3. Other results

There has been very intense activity in the atmospheric sciences over the last few years and it is difficult to give even a summary of the most recent work. The reader should therefore refer to the articles published in the scientific press, remembering that many of them have provided the basis for the official reports referred to in this document. The summary record of the meeting of the UNEP Coordinating Committee on the ozone layer (Paris, November 1979) describes the results obtained so far and the research being carried out in Canada, France, the Federal Republic of Germany, Italy, Netherlands, Norway, the USSR, the United Kingdom and the United States.

2.4. Differences between the American and British conclusions

The first point to be made is that the mathematical models used in both the United States and in Europe (particularly in the United Kingdom) provide, for similar assumptions, quantitative results which exhibit a high degree of comparability. The ozone depletion figures obtained by very similar computing techniques are compatible on both sides of the Atlantic.

The basic difference between the American and British approach lies in the confidence placed in the models and the degree of simplification considered to be acceptable. In the American case, the effect of perturbations calculated with the aid of one-dimensional models is taken to be representative of the real situation since these models are quite successful in simulating the natural atmosphere. In the British case,

the attitude is that atmospheric phenomena are highly complex, that it is therefore impossible to make such over-simplifications and that models having only one dimension - which cannot simulate a number of phenomena in the atmosphere (sudden warmings, variable height of the tropopause, effect of planetary waves, etc.) - are suspect when used to predict ozone depletion. Appendix 1 - included by way of illustration - sets out more explicitly the respective philosophies underlying the American and British studies.

In other words, it is the methodology which is called into question here and not the calculations themselves. In practice, the quantitative differences between the findings of the simplified British and American models are small and, in the last resort, of little significance. The key question is to know whether there can be any appreciable difference between the calcultated amount of ozone depletion and the actual response by the atmosphere to the effect of CFCs. Thus the problem is that of validating the models and the method of formulating the problem.

3. UNCERTAINTIES ARISING FROM ATMOSPHERIC MODELS

3.1. General

Both the American and British reports deal with the uncertainties and errors which might be introduced into mathematical models. The NASA report makes a clear distinction between the errors inherent in the formulation of phenomena by simplified equations and the errors due to a poor understanding of the input parameters. In the first case, the problems arise from the simplification of the equations; in the second case, from an imperfect understanding of atmospheric chemistry and the transport of chemical substances. Account will also have to be taken of the part played by the coupling of chemical, thermal and dynamic phenomena and the inadequacy with which they are represented.

3.2. Validity of the methodology underlying atmospheric models

From the strictly theoretical point of view, all the atmospheric parameters can be determined by the detailed resolution of a number of equations to which are added boundary conditions and adequate initial conditions. These equations relate principally to the conservation of moment, energy and mass, and to the concentration of each chemical species. There is a generally high-degree of coupling between all these phenomena - to which must be added the gas state equation -and hence they must all be treated simultaneously.

In practice, however, it is vital to introduce a number of simplifying assumption or to use approximative methods of resolution. Such simplifications will be directly dependent on the type of solution required. As an example, the basic equations about the atmosphere reveal phenomena whose time or space scale may vary considerably and is also governed by the spatial resolution of boundary conditions. Thus a representation of the general average circulation can be obtained only if the smaller-scale phenomena are "filtered out" but, if the equations are handled with sufficient resolution, a description can also be given of the behaviour of pressure or temperature systems at synoptic or local level. Usually, the interactions between the various spatial and time scales are not considered. In the general circulation models, therefore, no account is taken explicitly of the transport of ozone by the meteorological systems which may nevertheless play an important part. In this context, the problem of representing the tropopause is typical. The altitude of this interface between the troposphere and the stratosphere varies daily, even if the seasonal or annual average is well defined. The amount of ozone in the stratosphere - a direct function of the height of the tropopause -, is bound to be affected by meteorological fluctuations of this kind. Yet the global models are not suitable for stimulating the transport of chemical species by these processes. The sudden warmings at the end of winter in the polar stratosphere also have

an effect on the quantity and transport of atmospheric molecules. Their origin - still not fully understood - would appear to be in the interaction between tropospheric waves and the general atmospheric circulation. The theoretical findings take no account of these phenomena when the basic equations are simplified, however slightly. Similarly, when a discretization technique is used to resolve the system, the phenomena whose characteristic scale is below the calculation grid are filtered out; this may even affect any large-scale solution.

To conclude, there are already serious problems regarding the methodology underlying even the complex mathematical models.

Gross simplifications are introduced into the aeronomic models. The transport and temperature distribution parameters are specified and one has solves the continuity equations related to the chemical species the concentration of which it is required to know. Furthermore, the approach to the problem specially in the NASA and NAS reports is merely "one-dimensional", i.e. the only variations taken into account are those distributed along the vertical. In other words, such models presuppose a perfect uniformity of conditions of latitude and longitude (perfectly stratified atmosphere) and the values sought or specified are based on average conditions throughout the globe. Since, on average, the components of the wind are nil, the general average circulation does not come into the one-dimensional problem. Hence, the continuity equation for a given species is written as follows :

$$\frac{\partial \bar{n}_i}{\partial t} + \frac{\partial}{\partial z} \left(\overline{n'_i w'_i} \right) = \bar{P}_i - \bar{L}_i$$

where $\bar{n}_i$ is the average concentration at an altitude z and time t, $\bar{P}_i$ and $\bar{L}_i$ are the average rates of production and destruction of the species i respectively, $\overline{n'_i w'_i}$ is the average value of the vertical component of the constituent i. The bar signs over the symbols denote an

average value at any latitude and longitude; the prime symbols relate to the deviations from these means (fluctuations).

In order to resolve the continuity equation, the mean fluctuation must be specified in terms of average values and, in particular, the average turbulent flux must be known in terms of the average values of the other parameters. An expression of the following type is generally adopted, i.e. :

$$\overline{n'_i w'_i} = - K \left[\frac{\partial \bar{n}_i}{\partial z} + \frac{\bar{n}_i}{\bar{H}} + \frac{\bar{n}_i}{\bar{T}} \frac{\partial \bar{T}}{\partial z} \right]$$

where $\bar{T}$ is the average temperature and $\bar{H}$ the atmospheric height scale. The parameter K is a coefficient of vertical exchange or turbulence whose profile can not be calculated theoretically and must therefore be worked out empirically. In preactice, the K coefficient is calculated on the basis of the observed profile of constituants such as N_2O, CH_4 or even chemically-inert radioactive particles. Figure 1 shows a number of profiles currently used in the American and British models; the significance of variations, especially in the middle stratosphere will be seen. It will also be noted that all the models use the same coefficient of transport for all chemical species without such an assumption ever being justified. For example, the vertical profile of the K coefficient (Chang 1976) has been calculated from the average distribution of methane as observed by several researchers (Fig. 2). It will be seen that this distribution is highly dependent on a value for CH_4 measured by rocket at a height of between 40 and 60 km and that any error in this reading would significantly affect the value of K (z). Furthermore, it inevitably depends on the concentration of those chemical species such as OH which attack methane and whose distribution is poorly understood at present. When the average distribution of the nitrogen hemioxide N_2O is calculated using the transport coefficient deduced by this method, it is not possible to obtain the same distribution of this gas as is observed at mid-latitudes. But it is true

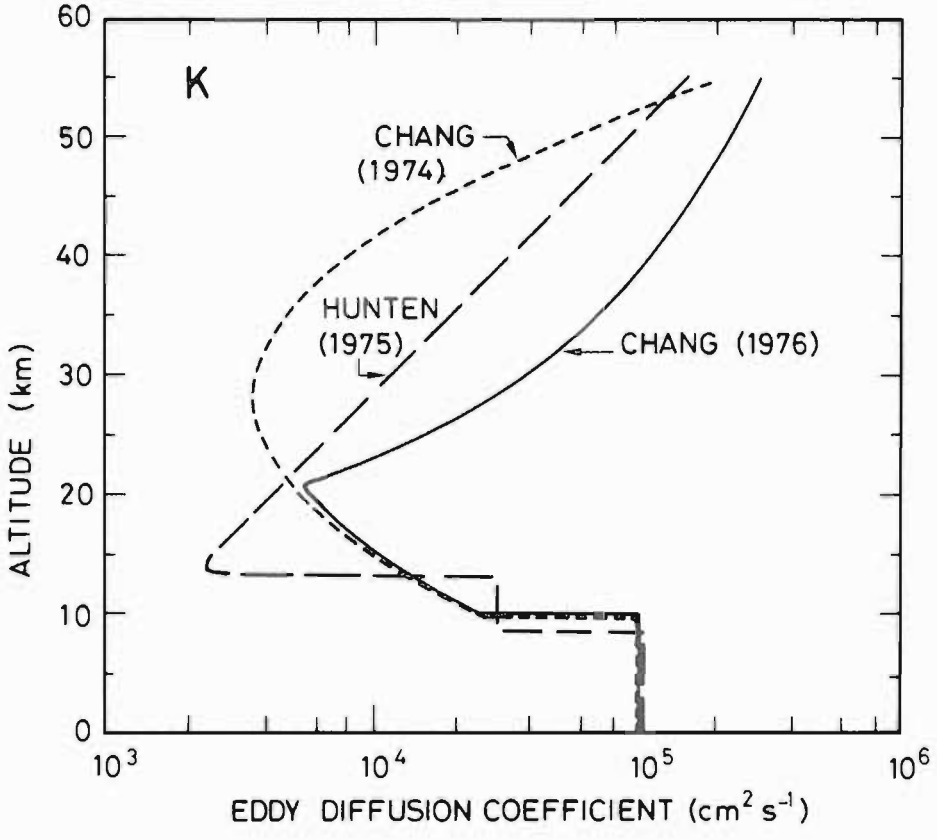

Fig. 1.- Vertical distribution of the transport coefficient used in one-dimensional models of the stratosphere.

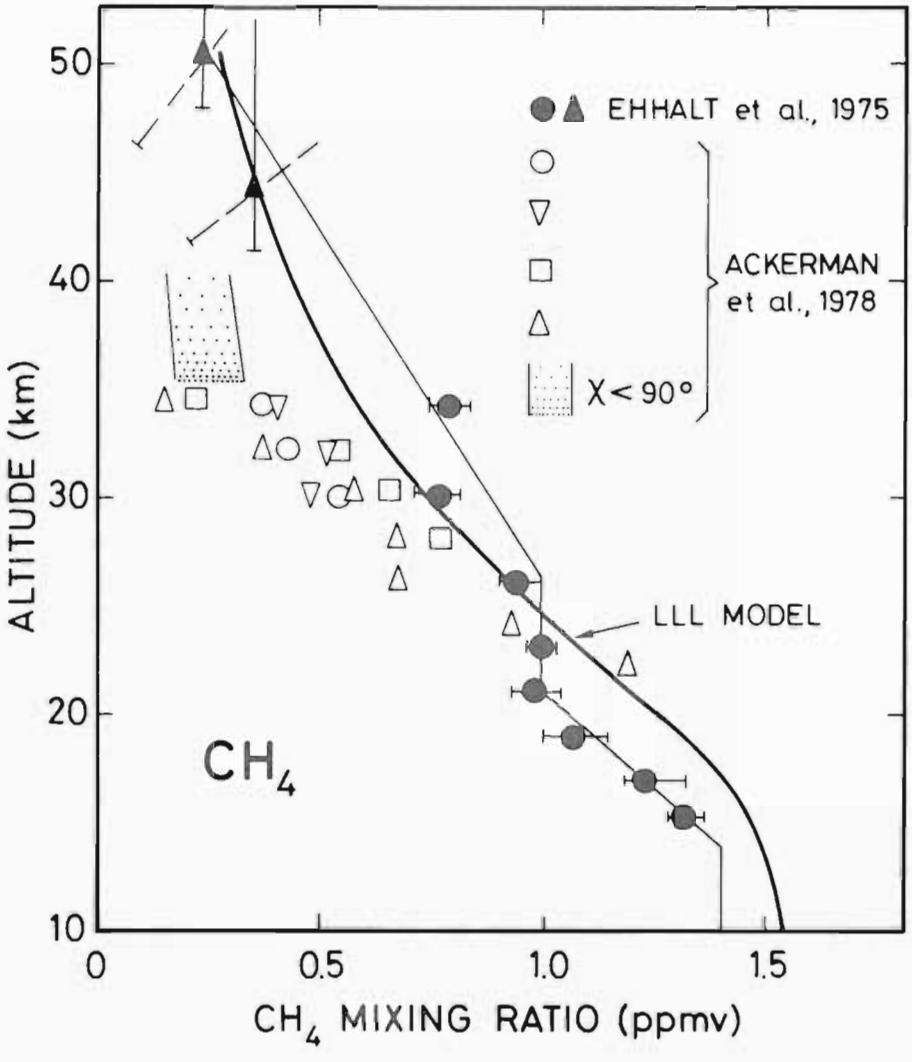

Fig. 2.- Vertical distribution of the volume mixing ratio of methane from which the value of the atmospheric transport coefficient has been calculated at all altitudes.

that beyond a certain level, the number of experimental data is inadequate to provide the basis for any definitive conclusion.

The two-dimensional models should lead to a more detailed representation of the distribution of chemical species but they do not enable any real progress to be made in the study of perturbations. Parameters in respect of the transport also have to be included and, basically the approach is similar to that of the one-dimensional models with the uncertainties greater. In practice, only highly complex three-dimensional models will bring any substantial progress.

To sum up, it must be said that - apart from the reservations one might have about the methodology relating to models in general -one-dimensional models are based on simplifications. Hence, this approach does not necessarily give a satisfactory reflection of the actual situation in the atmosphere, a point which the British report did not fail to stress. This report summarized the difficulties inherent in modelling by listing the simplifying assumptions generally made in the following way :

1. Reduce number of chemical species considered.
2. Reduce number of dimensions of the model.
3. Use climatological data rather than meteorological equations.
4. Limit spatial resolution.
5. Reduce time dependence.
6. Problems in respect of the assumptions used to calculate the average of the coefficients or the product of the concentrations.

Despite such varied assumptions it must be stated that, in general terms - and sometimes at the expense of certain adjustments - the distributions calculated are not inconsistent with the profiles observed. In other words, the natural atmosphere is fairly well simulated by these simplified models. But can the same be said for the perturbations of the atmosphere ?

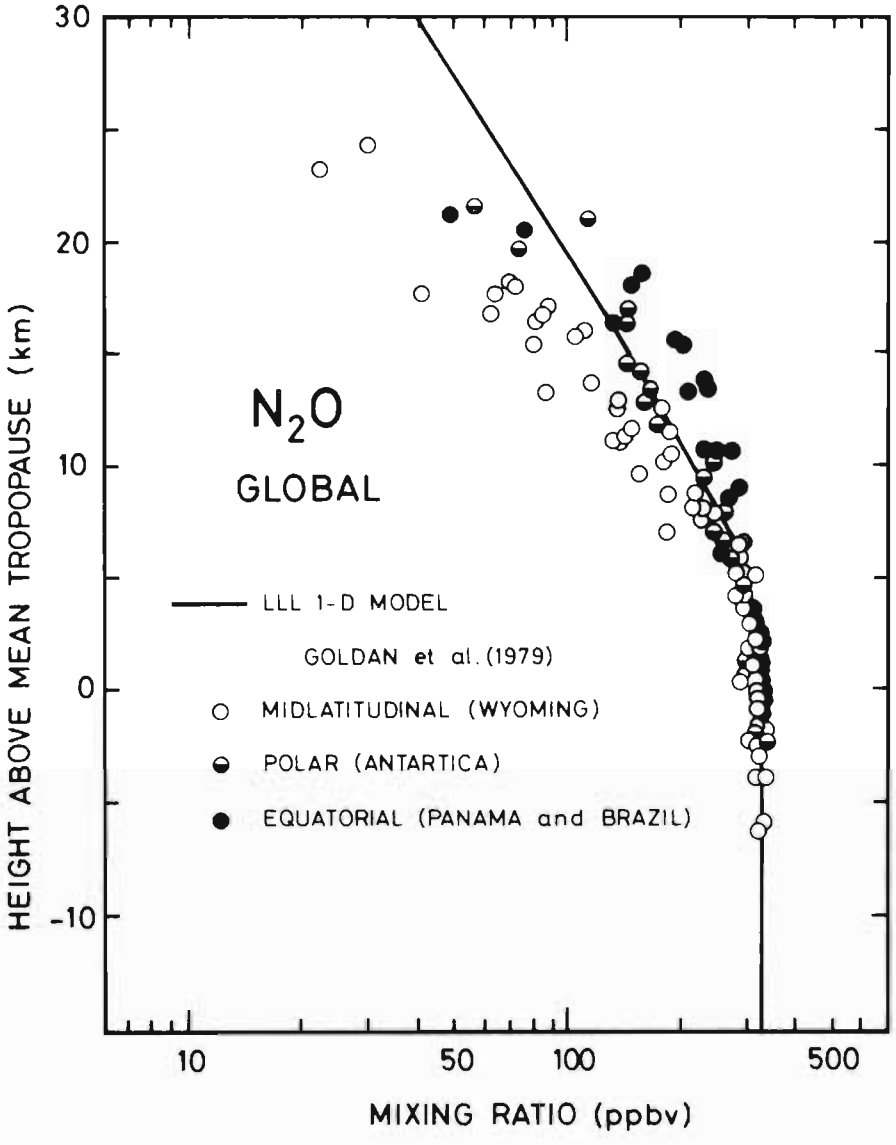

Fig. 3.- Comparison between the observed and computed values of the volume mixing ratio of nitrous oxide.

3.3. Uncertainties due to atmospheric chemistry

Our knowledge of the chemical and photochemical mechanisms which operate in the atmosphere has increased considerably in ten years. The rate of the majority of chemical reactions has been measured and re-measured in the laboratory during the 1970s. Yet a number of rate constants were determined using only one experimental method and should therefore be re-measured using other methods. Furthermore, the reaction products are rarely detected : this is also the case for photo-dissociation reactions induced by ultraviolet light. Moreover, the reactions of various species are rarely measured in a nitrogen/oxygen system comparable to that of atmospheric air. Indeed, not enough is known about the existing link between reaction rate and air pressure and measurements should be taken to remove this uncertainty.

The NAS report describes the progress achieved in this field in recent years, stating that a number of new findings have sometimes radically altered the estimate of ozone depletion caused by CFCs. As an example, the reaction of HO_2 with NO was re-appraised less than three years ago and the new constant has virtually doubled the CFC effect. The interaction between the hydrogen, nitrogen and halogen families is currently thought to be an important phenomenon. The NAS believes that present laboratory techniques are more sophisticated and will enable chemical reaction rates to be measured directly. The NAS report states that it is unlikely that major changes will occur in the future regarding the value of the reaction constants. This may be too optimistic since the latest major changes took place only quite recently in 1977 and 1978.

Since the publication of this report, a number of significant revisions in some reaction rates have been suggested : the rate of reaction $OH + HNO_3$ should be, according to Ravishankara (1980), 3.15 times faster at 225 K (lower stratosphere) than the value previously

accepted and should also be temperature dependent[1]. On the other hand, if the value recently proposed by Lii et al. (1980) and De More (1979) is accepted [2] the recombinaison of OH and HO_2 should be 2.5 to 5 times faster than recommended in the NASA report. It has been suggested that this important reaction rate could be pressure dependent but there is no scientific argument to explain this hypothesis. Furthermore, the rate constant of the reaction between OH and H_2O_2 could be 4 times larger at 225 K (Keyser, 1980; Kaufman, 1980) than the value recommended by NASA. These recent changes should reduce the ozone depletion calculated by the mathematical models.

Finally, serious doubts remain about components such as HO_2NO_2 or $ClONO_2$. Other examples of uncertainties could be given e.g. the products of reaction ClO + OH or ClO + HO_2, the photodissociation rate and products of several species such as NO_3, N_2O_5, HO_2NO_2, $ClONO_2$, HOCl, etc... It is interesting in this connection to show what has been over the years the consensus view on the level of ozone depletion either as a result of CFCs (Figure 4) or the oxides of nitrogen emitted by aircraft flying in the stratosphere (figure 5). In the first case, the observed effect has become more and more alarming over the years; in the second case, the observed effect -after having been very negative - is now reckoned to be marginally positive. The question therefore arises as to whether there are any reactions which have hitherto been overlooked and whose scale and effect (positive and negative) might become manifest at a later date.

To conclude, it would seem that much more is now known about the chemistry of the atmosphere than before but that unknown mechanisms may exist or reactions occur whose rate constant is over - or underestimated

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(1) $1.5 \times 10^{-14} \exp(649/T)$ cm^3 s^{-1} instead of 8.5×10^{-14} cm^3 s^{-1}

(2) $1\text{-}2 \times 10^{-10}$ cm^3 s^{-1} instead of 4.0×10^{-11} cm^3 s^{-1}

(3) $2.5 \times 10^{-12} \exp(-126/T)$ cm^3 s^{-1} instead of $1.0 \times 10^{-11} \exp(-750/T)$ cm^3 s^{-1}

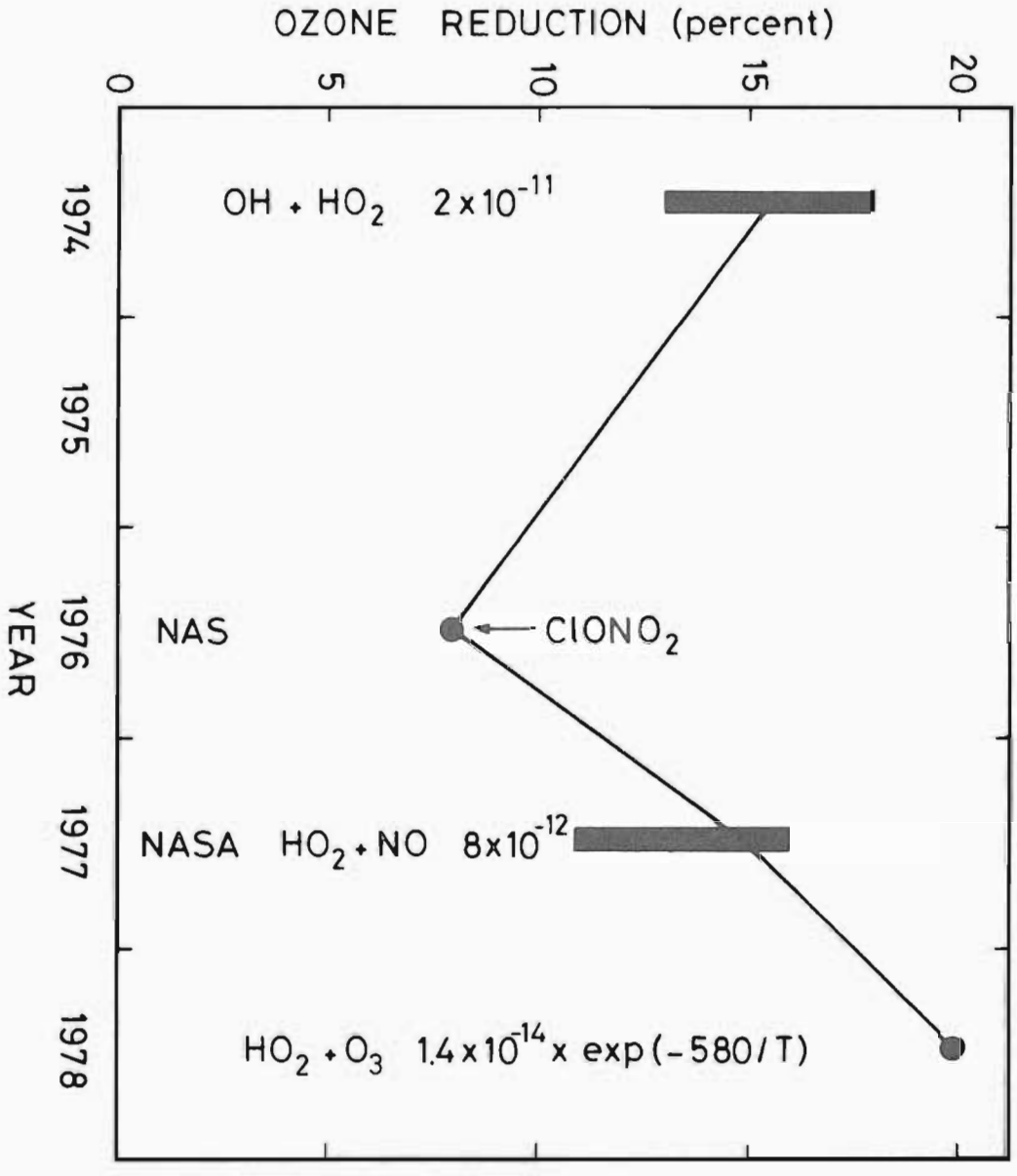

Fig. 4.- Principal modifications of the calculated destruction of ozone by chlorofluorocarbons. The modified parameters are shown (Ehhalt, 1978).

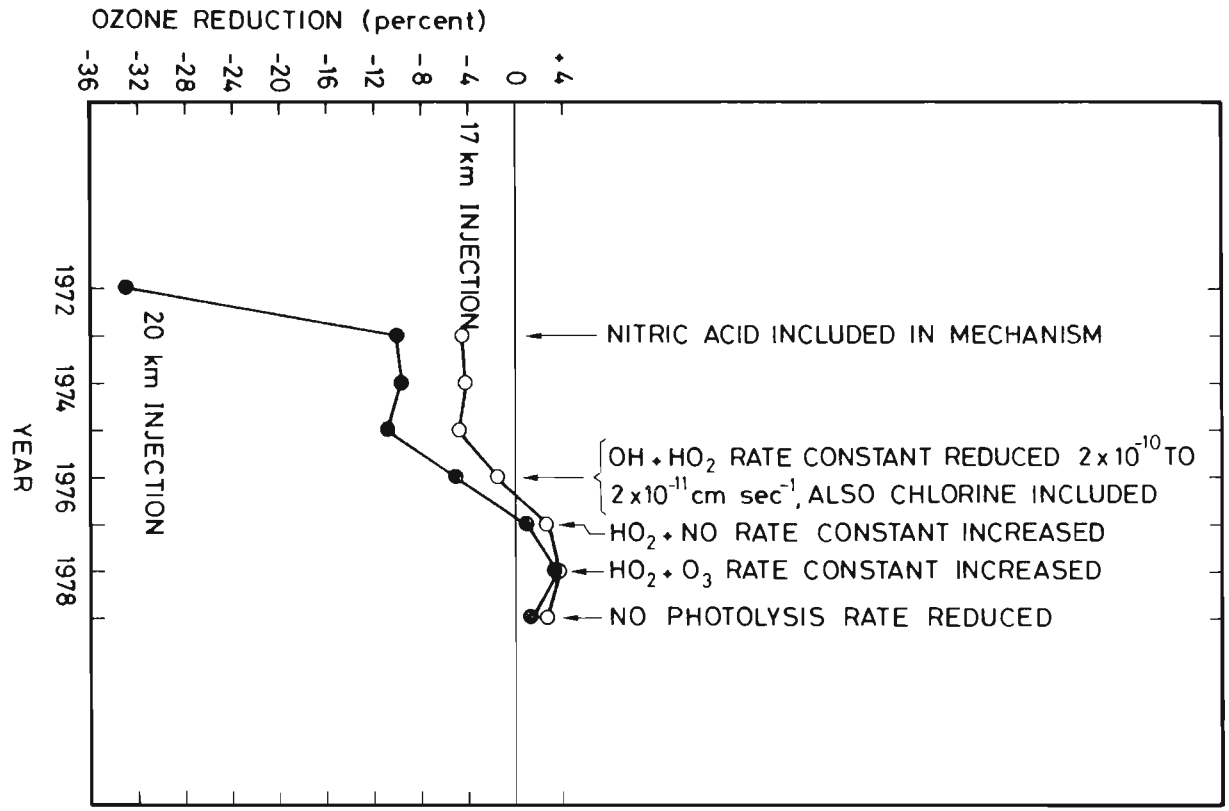

Fig. 5.- Principal modifications of the calculated ozone variation caused by a fleet of aircraft flying at 17 or 20 km altitude. Results obtained by the Lawrence Livermore Laboratory (LLL) model assuming an injection of 2 000 molecules cm^{-3} s^{-1} NO_x in a layer 1 km wide (Chang , 1979).

A certain degree of measurement error attaches to every reaction rate; the global error in respect of a system containing over one hundred reactions is quite considerable. This error has been estimated in the NASA and NAS reports and is the basis for the uncertainty limits stated earlier. It must be pointed out, finally that the chemistry of the troposphere is still imperfectly understood. More extensive research should be conducted, specifically, into the decomposition of CFCs in the troposphere and their possible disappearance into the oceans and soil.

3.4. Uncertainties in respect of solar radiation and its penetration into the atmosphere

Stratospheric ozone is produced by the photodissociation of molecular oxygen. Thus, to calculate the rate of ozone formation, it is essential to have a precise knowledge of the spectral intensity of the ultraviolet solar flux in the 180 to 280 nm range. Moreover, the ozone may itself become photodissociated notably through the action of radiation whose wavelength lies in the range 200 to 300 nm.

The spectral distribution of the solar flux has been measured by a number of investigators (see in particular the work of Simon and his report to the EEC) and tables of average values been drawn up for the benefit of model-builders. These values were recently called into question by Rottman as regards the spectral region in which the ozone is formed. Differences of more than 50% could occur. It is therefore fair to state that it is impossible at present to calculate - with the desired precision - the ozone concentration at an altitude of between 40 and 50 km, i.e. in the region where either (a) photochemical equilibrium is achieved and there is no direct transport effect or (b) where the chlorine-ozone interaction is greatest. Furthermore, since observations are infrequent at this height, making it difficult to make a comparison with the theoretical results, it is clear that even in an area where the conditions for the calculation are at their simplest, it is still not

possible to state with any certainty any average value for the rate of ozone production or even its concentration.

The way in which the penetration of radiation is treated, especially in the one-dimensional models, is often grossly simplified. Indeed, the molecular diffusion of light is dealt with in approximate fashion and more often than not the effect of solid particles and clouds is omitted. Similarly, the effect of the albedo and its variation in space and time is not considered. Lastly, the methods used to investigate the penetration of radiation in the Schumann-Runge bands of molecular oxygen lead to results with a certain degree of approximation.

3.5. Uncertainties due to the expression of the transport factor

It was stated earlier that the way in which transport was expressed in terms of parameters was directly linked to the type of model used. It has become clear that the method using the diffusion coefficient (K) was purely empirical. More especially, no proof is given that the same K profile can apply to all the species in the atmosphere. Similarly, there is nothing to prove that the theory of the "mixing length", which underlies the concept of the K coefficient, is valid in every circumstance particularly where there is an intense mean transport phenomenon.

This reservation about the very concept of large-scale eddy diffusion is hardly mentioned in the NAS report. It merely states that : "It is felt that the uncertainty in the eddy-diffusion coefficient at a given altitude is about a factor of 2, although it is admitted that this uncertainty estimate is somewhat subjective". The report goes on to say that, at all events, the prediction of ozone depletion under the effect of CFCs is not very sensitive to the value of the K coefficient.

The British report, on the other hand, shows that the ozone depletion is highly sensitive to the K profile chosen, as Table 2 - based on Dervent's calculations - shows.

TABLE 2.-

K profile	ClX at 50 km (10^{-9})	Ozone depletion (steady state)
Hunten	10.4	22
Chang 1974	9.8	21
Ehhalt	6.9	14
Chang 1976	5.9	12
Brasseur (max)	2.9	6

This dependency of the results on K is also clearly shown in Fig. 6.

If the problem is dealt with using a two-dimensional model (latitude, altitude) the parametric expression of the transport factor is more complex. The general average circulation is incorporated by the meridional and vertical components of the average wind value and the effects on atmospheric waves by an exchange tensor $\bar{\bar{K}}$. This is characterized (see the Reed and German theory, 1965) by the components K_{yy}, K_{yz}, K_{zy}, K_{zz}, which must be specified. Since these parameters are poorly understood, they often have to be adjusted by repeated trial and error method until the distribution of the chemical compounds in the meridional plane broadly corresponds to the pattern as observed. This approach leads to uncertainties of the same type as those referred to in connection with the one-dimensional models. Clough also demonstrated that the values obtained for ozone depletion by CFCs were heavily dependent on the choice of the K_{yy}, K_{yz}, K_{zy} and K_{zz} components. Moreover, Pyle recently questioned the very theory of representing the transport factor by the tensor K, pointing out that the theoretical value of the K components differed for each minor constituent and was thus a function of the chemical reactions to which these constituents were subject. For an inert gas in particular, the theory shows that

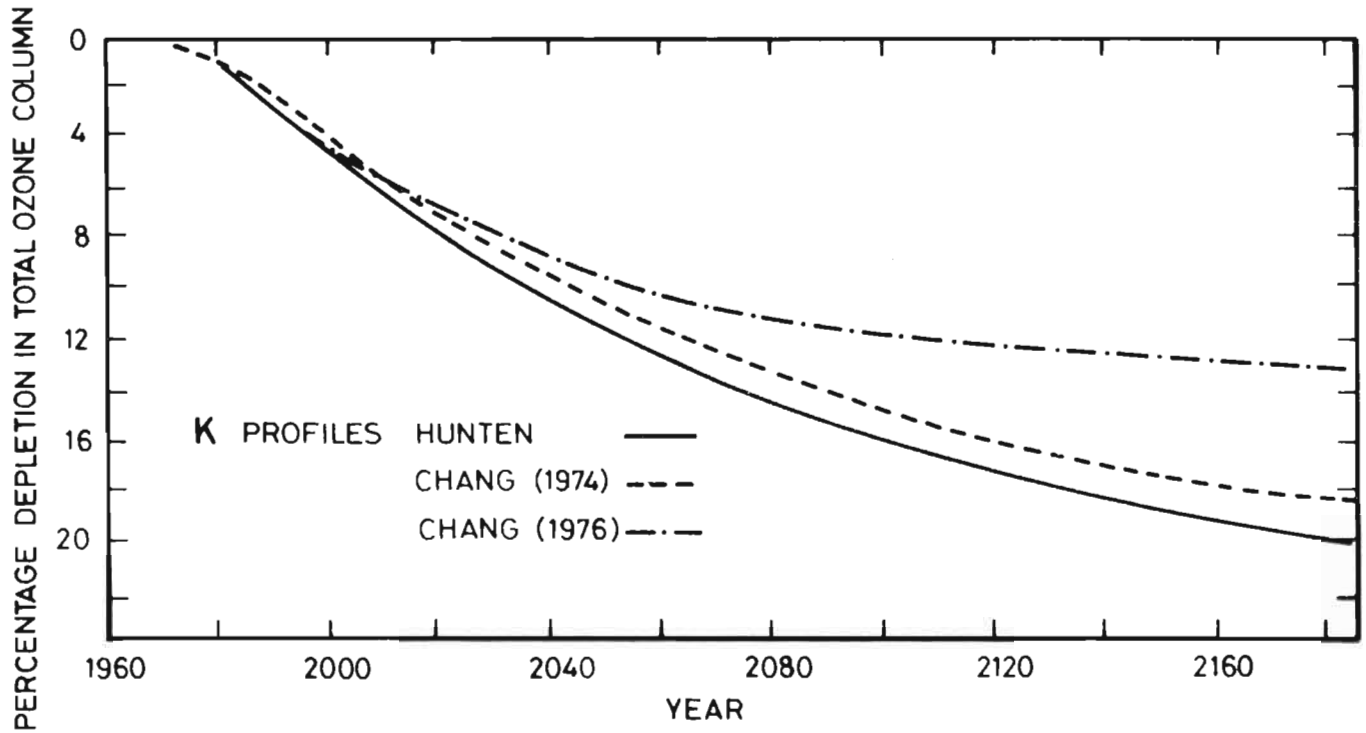

Fig. 6.- Trend in the depletion of the total quantity of ozone caused by CFC release (average production between 1973 and 1976) calculated by Derwent (Harwell-UK) in respect of different vertical transport (K) coefficients.

$K_{yy} = K_{zz} = 0$ and $K_{yz} = - K_{zy}$, this blatently contradicts the classical approach used by the two-dimensional models.

To conclude, no solution has been found to the problem of how to represent the transport of minor constituents. To do so will require the development of large-scale three-dimensional models necessitating complex calculations. The British report lays greater stress on the uncertainties due to transport than do the NAS and NASA reports.

3.6. Importance of coupling

The simplest mathematical models used to predict possible depletion of the ozone layer merely simulate the trend on the basis of chemical reactions, specifying other parameters such as transport or temperature. In actual fact, whenever the concentration of gases such as ozone or carbon dioxide varies, the rate at which the atmosphere is warmed or cooled is changed; the same is thus true for temperature, and hence for the dynamics. Certain models attempt to take account of those linkages which might regulate the atmosphere and thus give rise to effects markedly different from those induced by man. In the Pyle model, for example (Oxford, UK), the ozone depletion in 1990 is estimated to be 4.2% if no account is taken of the "dynamics" feedback and only 3.4% if it is allowed for.

The authors of the British report, however, lay emphasis on the complications which are caused by introducing different simultaneous perturbations such as :

(i) an increase in the CFC level
(ii) an increase in CO_2 due to combustion processes
(iii) an increase in N_2O due to the use of nitrogenous fertilizers
(iv) an increase in the amount of other gases such as CH_4, CO and NO_x.

By way of illustration, the British report shows that a 10% ozone reduction, caused in theory by CFC perturbation, drops to 7% if the amount of CO_2 is doubled; such an increase is expected in the next century. On the other hand, the increased use of nitrogenous fertilizers would, according to the British report, have an adverse effect on the ozonosphere but the effect of N_2O should not be "added" to the chlorine effect in view of the interaction between nitrogen and chlorine compounds.

3.7. Theoretical findings and observations

A comparison between the computed and observed distributions of minor constituents is an important aspect in model validation. Such a comparison - does not, however, provide absolute proof of a model's validity. On the contrary,

1. the observations of the majority of chemical species are very fragmentary, or need to be treated with circumspection;
2. the models provide average distributions which are not necessarily comparable with the values observed at particular points in time and in space;
3. the models may produce similar profiles for differently selected chemical or dynamic coefficients;
4. even if the theoretical findings are satisfactory in respect of the natural atmosphere, there is nothing to suggest that the effect of a perturbation as calculated by a simplified model would be representative of the real situation.

It is not possible in this brief report to deal with all the chemical constituents and to assess for each one the extent to which theory is borne out by observation and to what extent the observed data are valid. We will thus confine ourselves to a few typical examples.

The ultimate aim of the models is often to stimulate accurately the behaviour and hence the distribution of ozone. Distribution varies with latitude and season; the problem should therefore be dealt with on a two-dimensional scale at least (latitude and altitude). An average vertical distribution of O_3 can be defined, however, on the basis of all the observations and the one-dimensional model given the task of representing this average profile. Figure 7 compares the distribution as computed by Chang at the Lawrence Livermore Laboratory (LLL) with the distribution given by the average profiles observed by rocket and balloon (US Standard Atmosphere, 1976). The theoretical and experimental results tally; one might therefore conclude that the one-dimensional models have attained the major part of their objective. However this result may hide the real difficulties.

It is known that ozone distribution is governed by 3 factors, i.e.:

1. Production of ozone by photodissociation of molecular oxygen.
2. Destruction of ozone by oxygen, hydrogen, nitrogen and halogen compounds.
3. Transport of ozone especially in the region below 30 km altitude.

This prompts a number of comments :

1. Uncertainty remains in respect of the calculation of the production because of insufficient knowledge about solar flux in the 200 nm region and also in respect of the effective absorption layer in this spectral region.
2. In order to calculate the rate of ozone depletion a precise knowledge is required of the concentration of other constituents as O, OH, HO_2, HO, NO_2 and HNO_3, Cl, ClO, HCl etc. At present not enough is known about these distribution patterns.
3. Transport is difficult to simulate especially in the lower stratosphere where variations with space and time - associated with the tropopause for example - cannot be reflected in one-dimensional models.

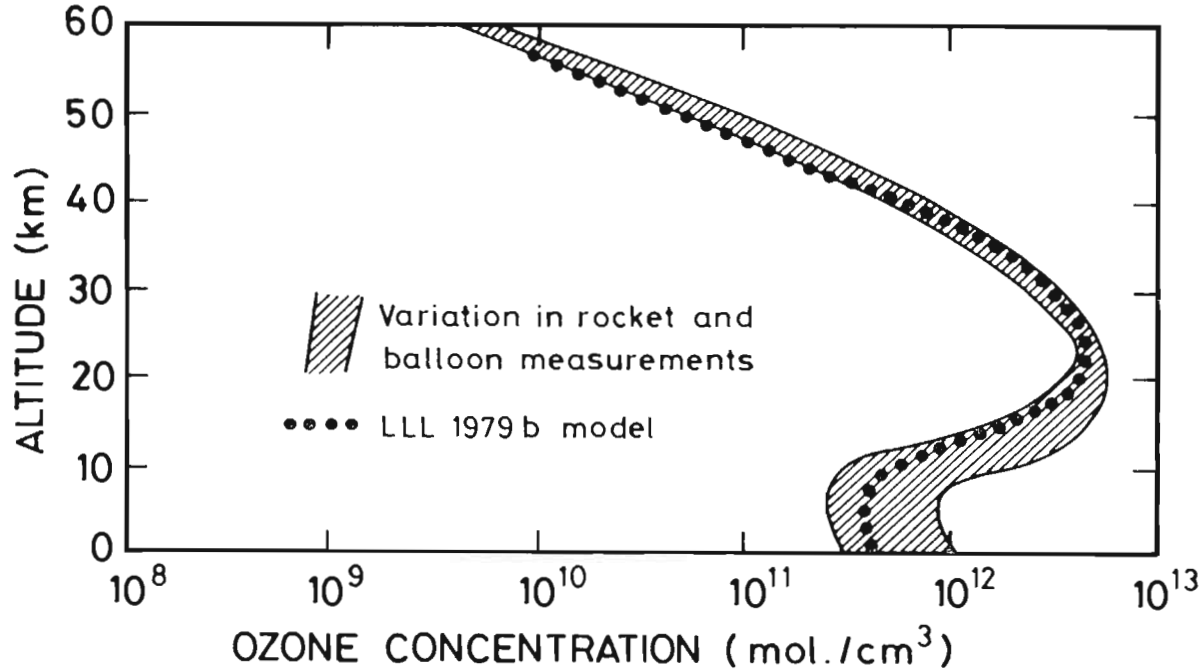

Fig. 7.- Comparaison between computed (LLL model - Chang 1979) and observed (US Standard Atmosphere, 1976) distributions of atmospheric ozone.

Whereas it may be said that by adopting the rate constants used in the NASA report vertical profiles for $O(^3P)$, OH, HO_2 can be obtained which tally more or less with current observed values, it must be pointed out that there are still problems to overcome when attempting to simulate the distribution of other species such as NO, HNO_3, ClO, etc. For example, figures 8 and 9 respectively illustrate the vertical distribution of nitric oxide and nitric acid. In the first case, it will be seen that the values calculated at mid-day are higher in the upper stratosphere than the values observed at the same hour of the day at mid-latitudes. In practice, the models usually overestimate the quantity of total nitrogen (NO_x), unless recent observations by Schiff in respect of the upper stratosphere were to be confirmed. As far as nitric acid is concerned, the higher computed values cannot be explained solely by the high concentrations of total nitrogen. These high values of HNO_3 could be explained by a gross overestimate for the concentration of OH but if the observed value of the radical is correct, this assumption must be rejected for the time being.

An analysis of the observed and computed distributions of ClO is especially important, the rate at which ozone is destroyed by chlorinated compounds being proportional to the concentration of this molecule. In the final analysis, therefore, it is this compound which provides the clue to the action of chlorofluorocarbons on atmospheric ozone. Figure 10 shows the observations made at mid-latitudes and the only observations made between September and December using the fluorescent resonance technique. It will be seen that observed and model values are in agreement at 40 km but that the latter do not correctly represent the slope of the ClO mixing ratio below this altitude. This discrepency may be explained in theory, by any of the following factors:

1. omission of an important chemical reaction in the theoretical calculation;
2. one or more incorrect chemical rate constants;
3. incorrect expression of the atmospheric transport parameters;
4. poor measurement technique.

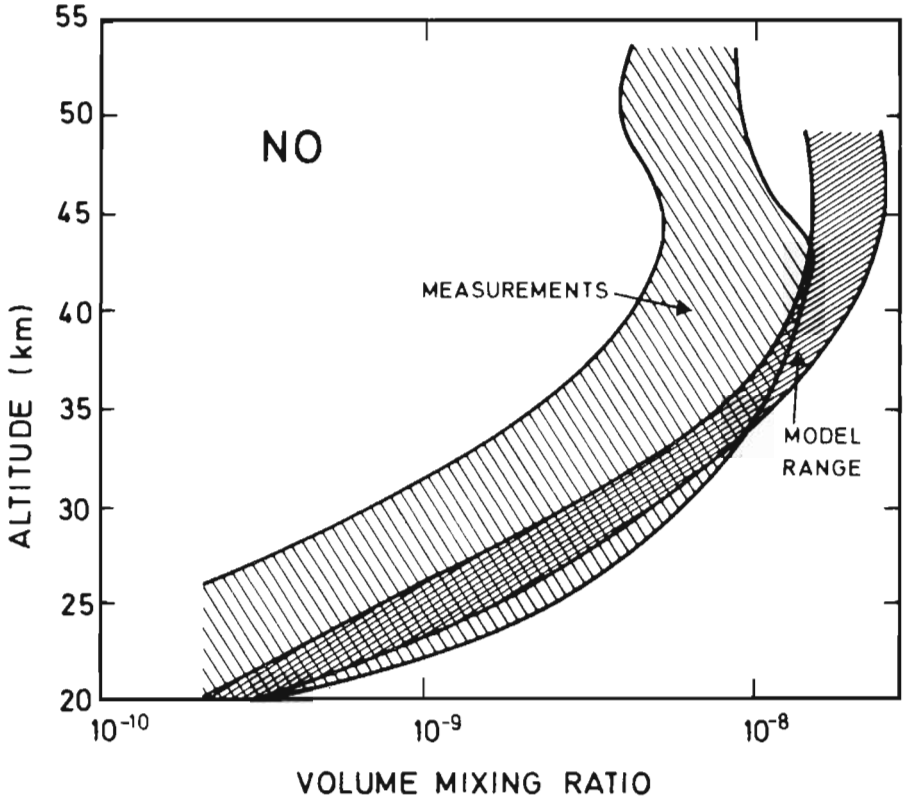

Fig. 8.- Comparaison between the observed and computed vertical profiles (American models) of nitrogen monoxide.

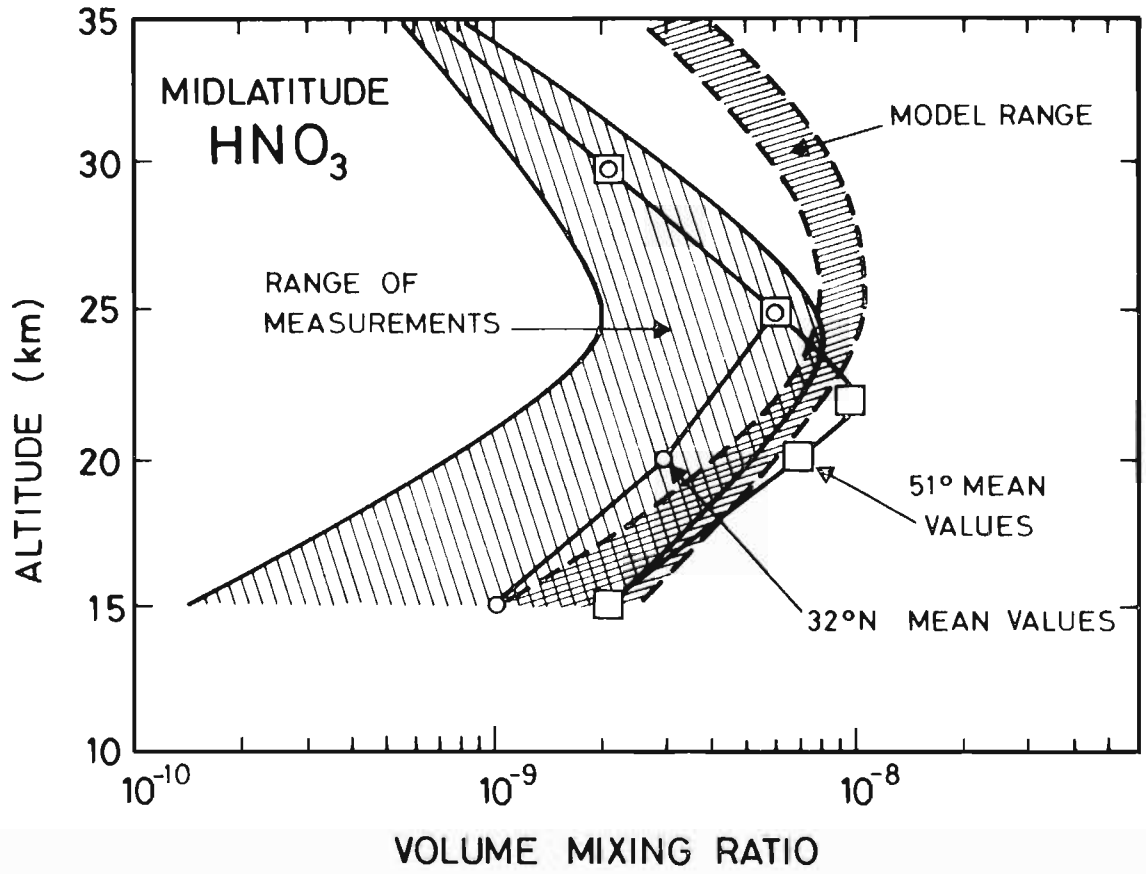

Fig. 9.- Comparaison between the observed and computed vertical profiles (American models) of nitric acid.

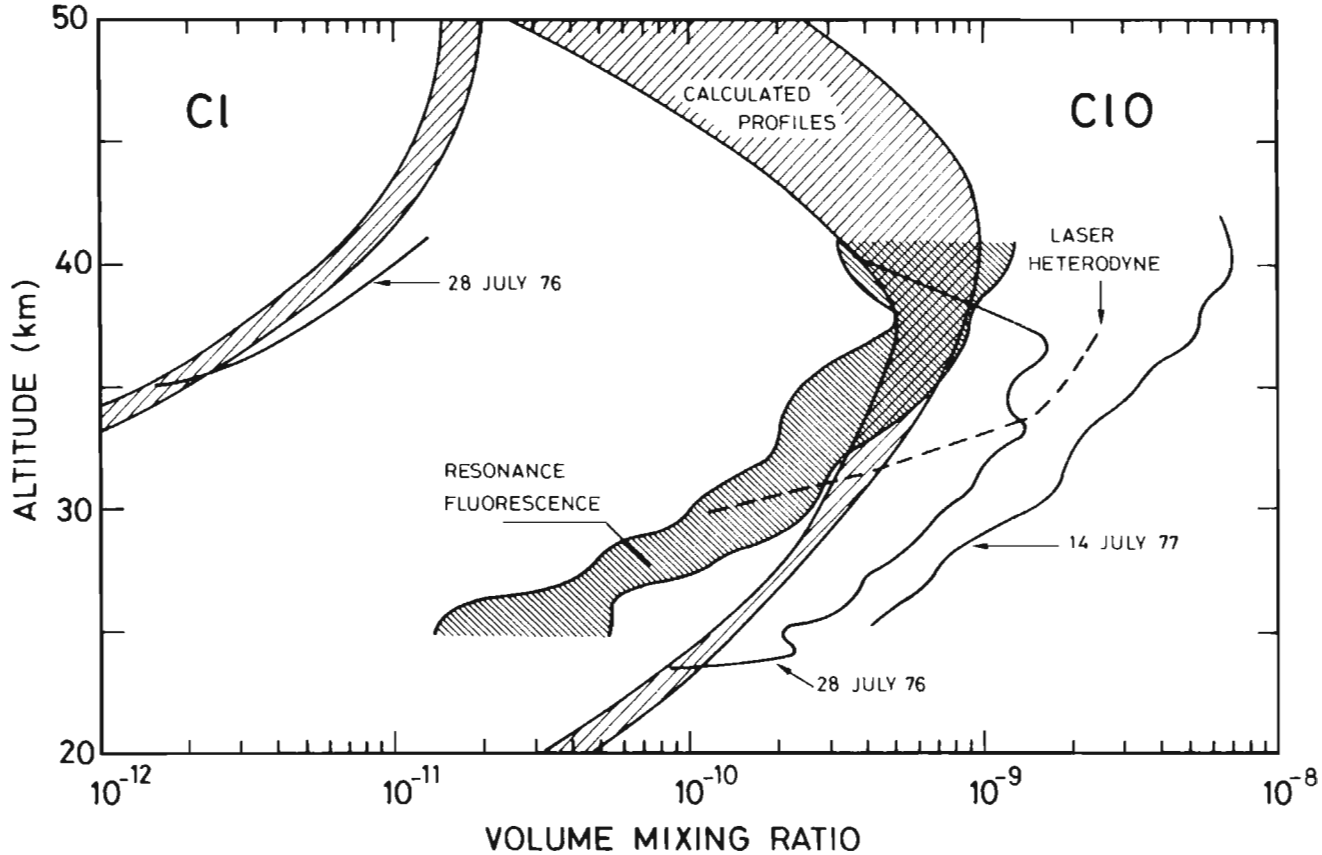

Fig. 10.- Comparaison between the observed and computed vertical profiles (American models) of chlorine monoxide.

The ClO observations made by the fluorescent resonance technique in July 1976 and 1977 (midday) and the measurements by heterodyne laser in September (sunset) give very high ClO readings. For example, the measurements taken by Anderson on 14 July 1977 showed a total quantity of ClX at 40 km equal to the values one would expect over several decades (a century) if CFC emissions continued at 1975 levels. It would therefore appear that atmospheric ClO is subject to substantial variations for which neither theory nor mathematical models have an explanation. Moreover, these large quantities of ClO do not appear to have any major adverse effect on the ozone layer, a phenomenon which Anderson attributes to one of the following arguments :

1. ClO has, prior to the measurement, been injected into the observation region over a period of time which is less than or equal to the response time of the oxygen combined in the form of ozone.
2. The reaction $ClO + O \rightarrow Cl + O_2$ is not the one which determines the rate of ozone depletion by chlorinated compounds.
3. Some ozone is also produced by chlorine; this counter-balances the ozone depletion and is thus of the same order of magnitude as the rate of formation of ozone by the photodissociation of O_2.

The classical theory of chlorine/ozone interaction does not therefore explain the present high concentrations of ClO observed and, if either Point 2 or Point 3 above were to prove correct, the estimates yielded by the models would have to be entirely revised.

To conclude this comparison between model and observation, it is worthwhile examining the indices used to validate the computed perturbations of ozone by CFCs. The molecules of CFC 11 and CFC 12 are a source of both chlorine and fluorine in the atmosphere. The HF/HCl ratio resulting from past production of CFCs can therefore be calculated. Figure 11 compares the theoretical findings of Derwent (British report)

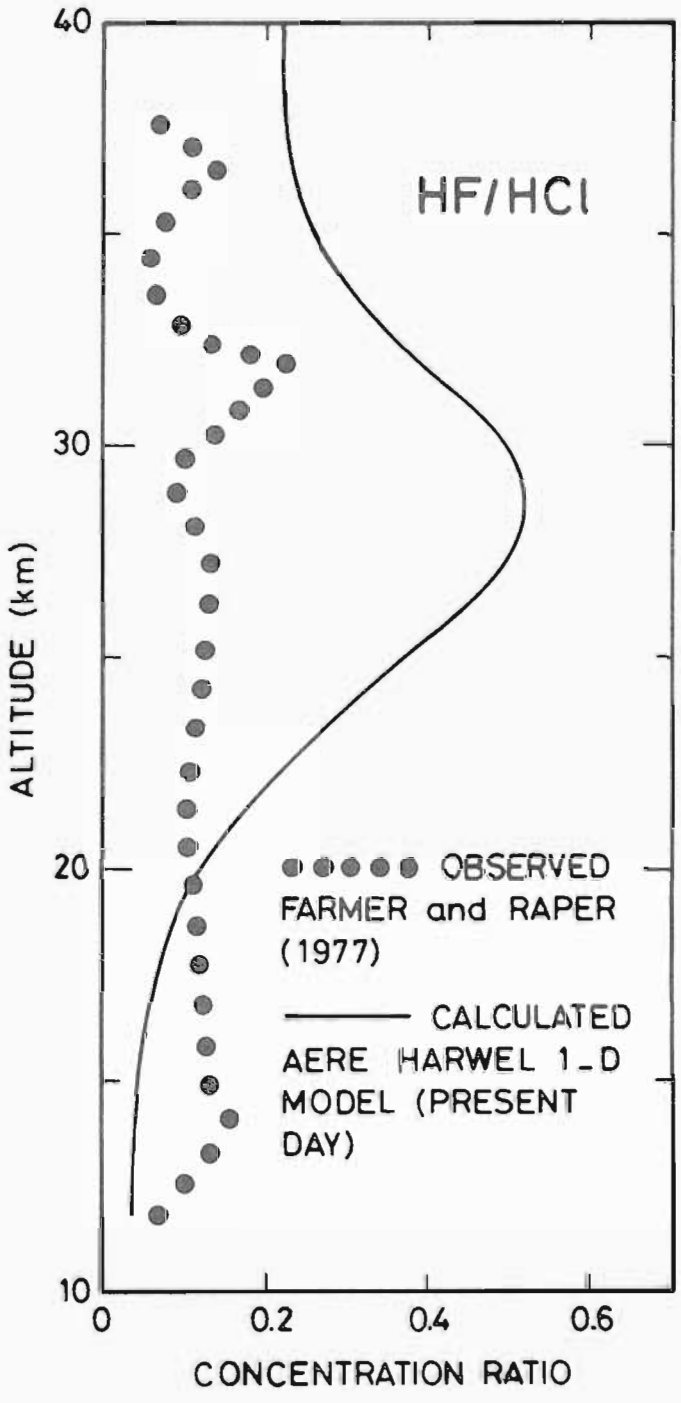

Fig. 11.- Comparaison between the observed and computed (Derwent) vertical profile of the ratio between HF and HCl concentrations.

with the observations made by Farmer and Raper (1977). There is considerable discrepancy between the results. In order to approximate to the theory underlying the observation in the upper stratosphere, one must postulate the existence of additional injections of chlorine. Perhaps the natural sources of ClX have been overlooked. If so, the computed ozone estimates would also need to be revised.

To conclude, even though the models have, in many respects, producted satisfactory results in simulating the natural atmosphere, uncertainties still remain, and these could significantly affect the estimates of ozone layer depletion of CFCs. Not only are there uncertainties in the basic data but methodology underlying the models should be treated with circumspection; furthermore the findings in respect of the natural atmosphere are not entirely satisfactory. A fortiori, no absolute trust can be placed in the quantitative results of the models which are used to predict ozone depletion.

3.8. Can more reliable results be expected in the next five years?

The foregoing analysis shows the need for further research. But if the requisite decision is delayed, the likely effect of CFCs may be greater and the consequences more serious. The British report shows that by extending the date of an assumed total cessation of CFC emission from 1 January 1979 to 1 January 1983, the maximum amount by which the total quantity of ozone would be depleted would increase from 0.5 to 0.6% since, according to the model, the interval between the cessation of emissions and the maximum O_3 depletion lies in the range 17 to 15 years. Consequently, a delay of 5 years before any decision is taken on CFCs can be reasonably accepted. But the question then is : What is the likelihood of fully resolving the problem within this period ?

Whereas an intensive research effort with adequate financial backing should help solve the problem as a whole from both the chemical

and meteorological points of view, it is our opinion that many more years and more effective computing techniques will be required in order to develop more sophisticated mathematical models (three-dimensional models, which take account of couplings etc.). An analogy may be made with the problem of computer weather forecasting where progress is significant but necessarily slow.

Although our understanding of aeronomic reactions and their rate constants can be expected to improve, the problems relating to the methodology of mathematical modeling will not be resolved within five years.

It would therefore be worth examining other means of estimating the effect of CFCs on ozone. In particular, monitoring the ozone layer and studying its trend should increase the likelihood of getting some idea over the next five years of the reaction of the ozonosphere to industrially-produced chlorinated compounds.

4. OBSERVATION AND TREND OF THE QUANTITY OF OZONE IN THE ATMOSPHERE

4.1. General

The quantity of ozone in the atmosphere and, in particular, its vertically-integrated concentration undergoes variations which follow different time-scales. Probe sampling shows that the ozone undergoes variations throughout each day under the influence of the meteorological conditions (synoptic situation, height of the tropopause etc.). There is also a very marked annual variation, the amplitude of which is no greater than that of the sporadic variations connected with the meteorological conditions. If all these phenomena are filtered out, there remains a small variation which may be caused by any number of factors : quasi-biennial oscillation, the solar cycle, human activity etc. Fig. 12 shows the relative trend of the total quantity of zone in several areas

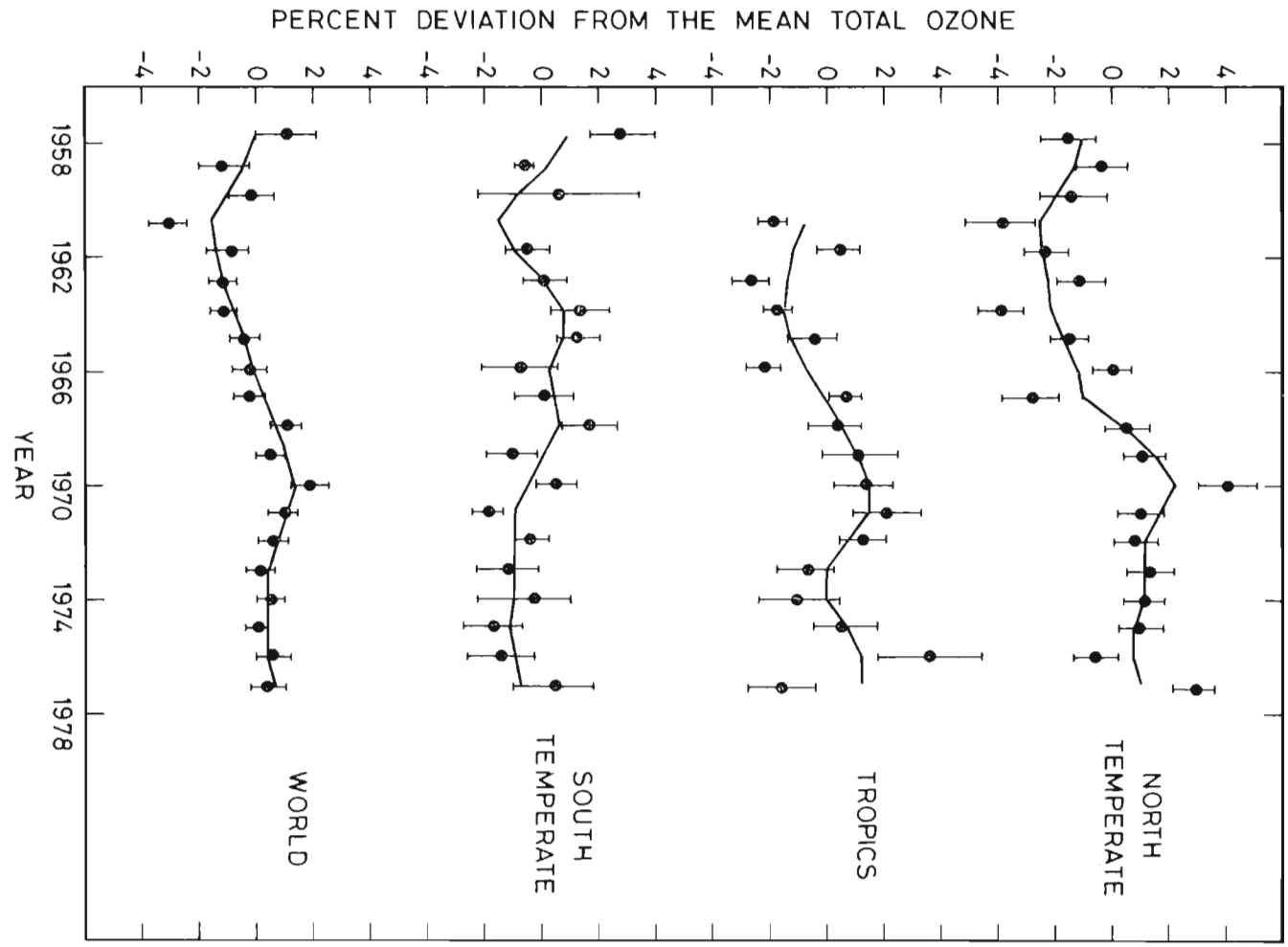

Fig. 12.- Trend in time of the variation in the quantity of ozone (annual average) compared with this average between 1958 and 1977. The vertical bars represent a 95% confidence level (NASA Report, 1980).

and the average for the world as a whole. In the temperate latitudes of the northern hemisphere, the quantity of ozone would appear to have fallen until 1962, rising to a peak in 1970 (+ 4%). There has been very little variation since then, although the quantity of ozone has fallen slightly. In the tropics, the average curve is more or less parallel to that for the northern hemisphere but in the southern hemisphere very different variations have been recorded.

4.2. Threshold of detectability

Is it possible to detect with certainty from curves similar to those in Fig. 12 any effect the CFCs may have had ?

To answer this question, it would be worth remembering what, according to the models, the present reduction in the quantity of ozone should be.

According to the NAS report, the action of CFC 11 and 12 should lead in 1980 to a reduction of 2.1% ± 1.5% of the quantity of ozone. To this figure should be added the effect of the other CFCs and in particular CH_3CCl_3 and CFC 22 (approximately 1%).

According to the British report, the present reduction in ozone due to CFC 11 and 12 is only 0.7 to 1.3%, depending on the model.

The NAS considers that it is impossible to detect with any certainty a variation in the quantity of ozone of less than 1.5% on a purely statistical basis. Takin into account other errors caused by the calibration of the Dobson apparatus, the uneven distribution of observation stations, meteorological and chemical effects upsetting the measurements etc, it would be difficult, in the opinion of the NAS, to attribute to CFCs a reduction in total ozone of less than 5% for 20 years.

The NASA makes a detailed analysis of all the possible causes of error and concludes that a global variation over 10 years must exceed about 6% if it is to be attributed to the action of CFCs.

The British report points out that, with the network of stations available at present and taking into account the number of independent observations made, it is difficult to attribute to CFC an observed reduction in total ozone lower than 2% per decade.

A recent study carried out in France (Nouyrigat, private communication) challenges these thresholds which are considered too high. This analysis attempts to show that it would be possible to detect a CFC-induced variation in the total ozone of 3% in the northern hemisphere and 2% in the southern hemisphere.

As for the observation of the ozone concentration in the upper stratosphere (40 km), the detectability threshold in respect of a CFC effect depends on the method of measurement used. According to the NASA report, these methods are as follows :

- rocket probe : 18.4% per decade
- Umkehr method : 15.6% per decade
- satellite : 5% per decade.

The majority of observations available over a sufficiently long period are based on Umkehr measurements but satellite data should provide more reliable results in the future.

The analysis carried out in France shows that - for the period 1960 to 1979 - it should be possible, using the data available (Umkehr), to detect a CFC-induced variation in the upper ozone layer of the order of only 2%.

On the basis of the NAS or NASA figures, therefore, it would seem that it is not yet possible to detect the effect of CFCs as shown by the models. If the British and, in particular, the French figures are accepted, the behaviour of the ozone should reveal a CFC effect and thus validate or disprove the mathematical models.

4.3. Total ozone

However, looking at the trend in total ozone for the northern hemisphere since 1958 (fig. 13), it can be seen that there has been a regular increase on average but with fluctuations which may be due in part to solar activity. Since several phenomena - either natural or induced by man (both known and unknown) - are involved and the measuring instruments may have deviated somewhat (or have been recalibrated) over the years, it is still not possible, in our opinion, to detect with any certainty an effect of CFCs on ozone.

4.4. Ozone in the upper stratosphere

In an attempt to achieve greater sensitivity, it is worth examing the observations of ozone concentration in the upper stratosphere. The photochemical theory indicates that the sensitivity of ozone to chlorinated compounds in this zone of the stratosphere is about three times greater in relative terms than the sensitivity of the whole body of ozone. The present reduction of O_3 at around 40 - 45 km would therefore be of the order of 5 - 9% depending on whether the British or American figures are adopted. These values should be compared with the thresholds of detectability. The Umkehr observations (figure 14) in fact show an increase of the order of 8% since 1965. There is therefore a glaring discrepancy with the elementary theory in respect of chlorine and ozone if one accepts the threshold given in the French study.

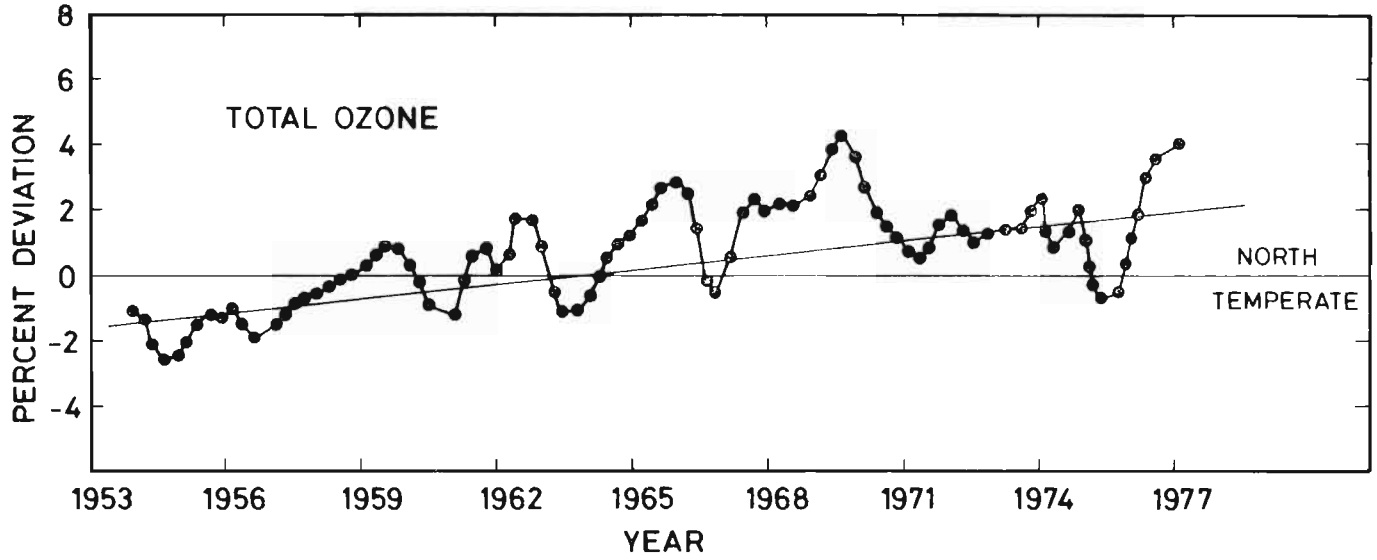

Fig. 13.- Trend in total ozone at mid-latitudes in the Northern Hemisphere from 1953 (Angell and Korshover, 1978).

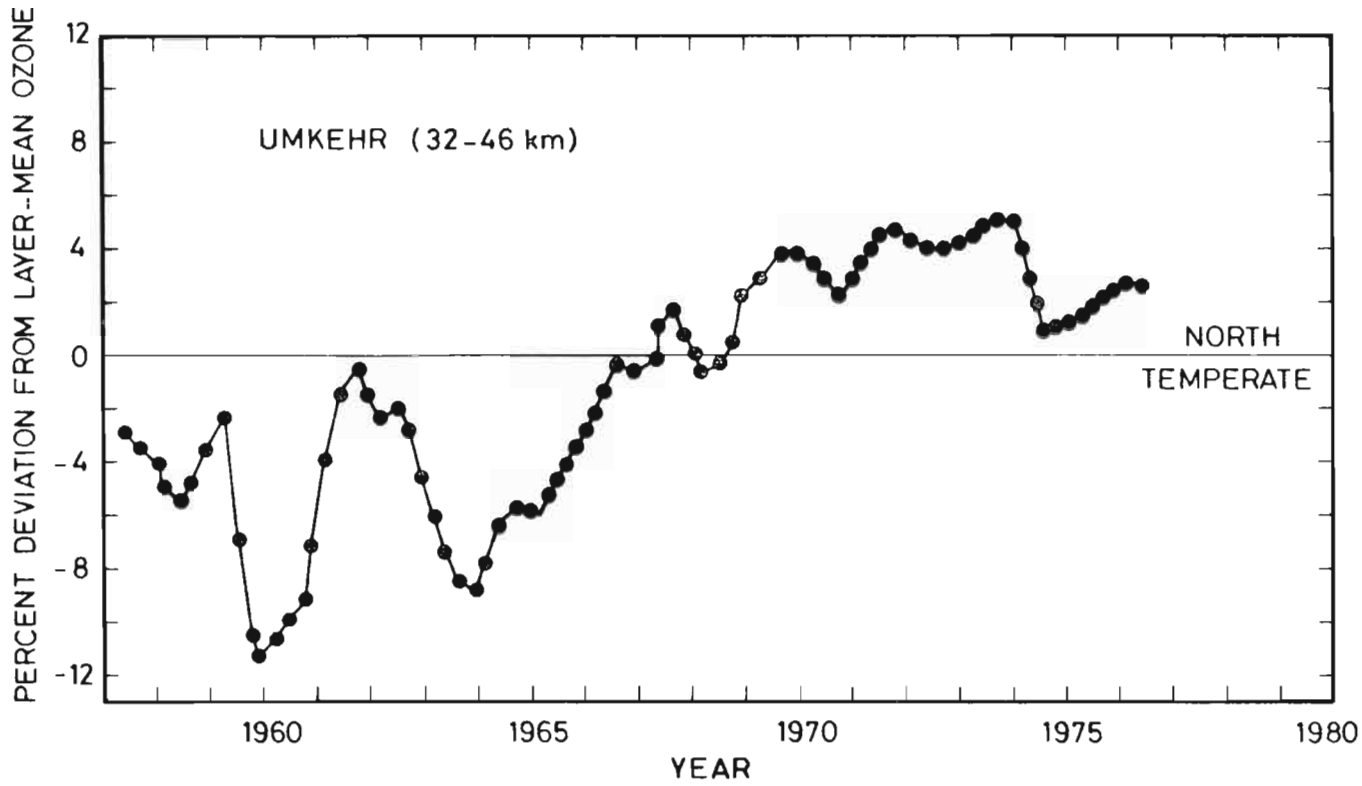

Fig. 14.- Trend in the quantity of ozone (seasonal average) in the layer located between 32 and 46 km at temperature latitudes in the Northern Hemisphere since 1957 (Angell and Korshover, 1978).

If, on the other hand, one accepts the threshold put forward by NASA (Umkehr), the CFC effect should not yet be visible. At all events, it will be the data from satellites which provide the first clue to any CFC effect. The observed increase in ozone can probably be explained by other phenomena which mask the effect of CFCs. Solar activity, in particular, whilst having some influence, is unlikely to be responsible for this ozone behaviour for, if the theory is correct, it should have helped to reduce the concentration of O_3 at 40 km since 1970.

In conclusion, the observations made and the analysis of them provide no clear proof of any action by CFCs on ozone. However, as the statistical analysis involved is by no means easy and there may be more significant phenomena in addition to the effects being investigated, and as the data sample available contains some measurements which may be wrong, caution must be exercised in making any final conclusion.

5. SUMMARY AND CONCLUSIONS

The recently published reports (NASA, NAS, HAPP, UNEP, etc.; see Table 1) have highlighted the probably effect of chlorofluorocarbons on atmospheric ozone. These conclusions are the outcome of intensive research carried out by the international scientific community since the time the hypothesis of the catalytic destruction of ozone by chlorine was first put forward by Stolarski and Cicerone (1974) and Molina and Rowland (1975).

Since then, the theory underlying this hypothesis had never been called into question. On the contrary, laboratory studies have shown that, in theory, ozone is sensitive to chlorine and the reactions which have been put forward since that time have merely backed up the theory without questioning its underlying principles.

Recent measurements of specific chemical reaction have, however, modified the quantitative values of the results obtained by the models and it has been shown that ozone is more sensitive to chlorine and less sensitive to the oxides of nitrogen than was previously thought.

Estimates of this kind are based on mathematical models which, in general, take into account a large number of chemical and photochemical reactions in the atmosphere but which are, at present, unable to simulate correctly the important role played by atmospheric dynamics.

The problem of the validity of the models thus takes on special importance since there is nothing to prove that the chemical mechanisms are at present understood.

Nevertheless, the models are very useful since they provide a better understanding of the relative importance of the various chemical and photochemical mechanisms and enable a study to be made of certain interactions taking place within the atmospheric system. However, the atmosphere is complex in this respect and caution must always be exercised when gathering quantitative results.

The British researchers, in their report, stressed the complex nature of the atmosphere and its meteorological mechanisms and consider that, from a scientific point of view, it is not at present possible to put a global figure on the ozone depletion caused by CFC emissions.

The American scientific community, on the other hand, with some ten mathematical models at their disposal, currently puts the level of total ozone depletion at 16.5 ± 10% assuming CFC emissions to the atmosphere at 1975 levels. These calculations also show that the present rate of O_3 depletion should be in the region of 3%, i.e. a value closer to the threshold of detectability.

An investigation of the actual trend in the quantity of ozone in the northern hemisphere (where the majority of CFC emissions occur) since 1963 reveals a positive tendency, the origin of which is not known. At present, therefore, the observed pattern does not tally with the theoretical results but one must not forget the possible combination of several natural (thermal, dynamic, chemical) effects or effects artificially created by man which conceal the direct action of CFCs.

It is worthwhile, in concluding this short analysis, to restate the main conclusions contained in the body of the report.

1. Mathematical models

Apart from the reservations one might have about the methodology relating to models in general, it must be said that the one-dimensional models lead to over-simplifications. Hence this approach does not necessarily give a satisfactory reflection of the actual situation in the atmosphere. Despite such varied assumptions, it must be stated that, in general terms - and sometimes at the expense of certain adjustments - the distributions calculated are not inconsistent with the profiles observed. For certain chemical compounds, however, a comparison between theory and practive does create certain problems. Uncertainties therefore remain which could substantially affect the estimates of the likely depletion of the ozone layer and the effect of CFCs.

2. Atmospheric chemistry

Much more is now known about the chemistry of the atmosphere but unknown mechanisms may still exist or reactions occur whose rate constant is over or under-estimated. A certain degree of measurement error attaches to every reaction rate and the global error in respect of a system containing over 100 reactions is quite considerable.

This error has been estimated in the NASA and NAS reports. Furthermore, the chemistry of the troposphere is still inadequately understood. More extensive research should be conducted, specifically, into the decomposition of CFCs in the troposphere and their possible disappearance into the oceans and soil.

3. Atmospheric transport

The problem of how to represent the transport of minor constituents has not been solved. This will require the building of large three-dimensional models involving advanced computer techniques.

Stated briefly, the conclusions are as follows :

1. There is now much more information available about the photochemical theory of ozone in the stratosphere than there was ten years ago;
2. Some uncertainties still remain mainly in the lower stratosphere;
3. The models have helped to improve our knowledge of the stratosphere;
4. Since the models are simplified, they cannot fully describe the behaviour of the atmosphere and its minor constituents;
5. In the next few years, more sophisticated models must be developed which can take into account simultaneously the chemical, thermal and dynamic aspects of atmospheric processes;
6. This is a task which cannot be completed within five years but steady efforts must be made in this direction;
7. Permanent observation and monitoring of ozone is therefore particularly important;
8. At present there is nothing to prove that CFCs have had a real effect on the ozone layer;

9. Observation facilities should therefore be developed, i.e. both satellite measurements, which supply a large number of observations, and ground measurements, which are easier to calibrate;
10. The examination of the balance sheets of the minor constituents should be continued in order to detect natural and artifical sources or sinks of these compounds in the atmosphere;
11. It is vital to study simultaneously all the effects of human activities on atmospheric ozone.

The problem of ozone and its vulnerability to compounds of human origin has now become a permanent problem. The figures now advanced will have to be revised frequently to take account of the developments of knowledge, the degree of sophistication of the models and the observations of the trace species.

APPENDIX 1

By way of illustration, the following parallel text sets out the conclusions of the report by the National Academy of Sciences (USA) and the report of the Central Directorate on environmental pollution (UK).

NAS report	UK report
"There is agreement with previous reports that continued release of halocarbons into the atmosphere will result in a decrease in stratospheric ozone".	"The validity of the hypothesis is still in doubt". "Basic scientific understanding, although progressing rapidly, is still inadequate in many respects". "The report concludes that present understanding of ozone depletion is limited and is based on model assumptions which have not been adequately validated".
"New values for some of the chemi cal rate coefficients have in creased the predicted ozone re duction resulting from continued release of chlorofluoromethanes (CFM's)".	"The results concur with other studies that the predicted reductions in total ozone amount are greater than those estimated in 1975 at the time of the preparation of Pollution Paper No 5. Whether the statements realistically describe what is likely to happen in the atmosphere depends on the validity of the calculations and also their coverage

	including the simultaneous effects of other atmospheric pollutants".
"The uncertainties in the chemical rate coefficients, in atmospheric transport, and in the use of one-dimensional models have been combined to give an overall uncertainty range of a factor of 6 within a 95 percent confidence mode level".	"It is not, therefore, realistic to assign overall uncertainty limits to our calculated ozone perturbations; deficiencies in our basic knowledge of the processes establishing the composition of the stratosphere and in the modeling technology cast doubts on their validity".
"The uncertainty range means that for the case of continued release of CFM's at the 1977 level, there is 1 chance in 40 that the ozone depletion will be less than 5 percent and 1 chance in 40 that it will be greater than 28 percent".	
"There have been considerable improvements in the computer model and in the laboratory and atmospheric measurements, which have reduced the uncertainty range".	"The STRAC report (The UK Stratospheric Research Advisory Committe) deals extensively with the uncertainties in the model results. Not all of them could be assessed quantitatively and it is not possible to assign error ranges to these estimates that allow for all the uncertainties. These have, however,

widened rather than narrowed since Pollution Paper 5 was published (1976)".
"It is evident that calculation of ozone reductions is made a more complicated exercise by these important feedback processes between photochemistry, radiation and dynamics, by no means all of which can be included in the present generation of models.

In considering possible modifications to the ozone layer an aim should be the development of multidimensional circulation models including complete treatment of all possible feedback mechanisms".

"Even allowing for the best pro fessional judgment of the possi bility that some important che mical reaction has been over looked or that there remain large errors in the measured chemical rate coefficients, we believe that there is a 3-out-of-4 chance that continued release of CFM's at the 1977 level will result in an ozone depletion that lies in the range of 9 to 24 percent".

"Any uncertainty analysis would be incomplete as it would encompass only quantifiable sources of uncercainty".

"In contrast, the uncertainty from the different assumptions made in simulating the transport processes and in the modeling procedures cannot yet be quantified, nor can any firm statement

	be made regarding the uncertainties arising from possible deficiencies in our knowledge of the detailed chemistry and photochemistry".
"Although there are a few exceptions, the comparison between the models and measurements of substances in the present stratosphere is considered to be satisfactory within the uncertainties of the measurements. We, therefore, believe that the projections for ozone depletion are valid within the stated uncertainty range".	"A certain amount of success in the simulations has clearly been achieved both in the STRAC programme and elsewhere, but there are major discrepancies. Moreover, unless one is prepared to reject the evidence of actual measurements in favor of the theoretical calculations, it must be conclude that there are still important gaps in our understanding and knowledge of the processes that determine stratospheric composition. It follows that if we are not satisfied with our present ability to simulate the natural stratosphere, particularly on the grounds that there may be major errors or omissions in the current theory, we must be very cautious in accepting quantitatively any predictions from accompanying perturbation experiments.

"The findings, together with other discrepancies between model calculations and measurements, bring into question the validity of the models presently used to predict ozone perturbations".

APPENDIX II

Conclusions of the research carried out by the UNEP Coordinating Committee on the ozone layer (Paris, November 1979).

The considerations leading to the prediction of an ozone depletion due to anthropogenic emissions of halocarbons like CFMs are plausible and largely consistent. To a large extent the models are consistent with measurements of atmospheric constituents, although discrepancies and uncertainties still exist. There is some difference of opinion between the delegates concerning the degree of confidence, which can be attached to the predicted ozone depletion, given the highly complex and partly understood processes occurring in the real atmosphere. Recent developments in statistical methods shows that a trend attributable to anthropogenic sources in total ozone of the order of three to six per cent is the smallest that could be detected by the present monitoring network. Within these limits, the present data do not show any significant trend in the total ozone amount. Model calculations supported by measurements remain the only tool for predicting future impacts of such activities.

Current models concur in a range of evaluations of ultimate ozone depletion with a most probable value of 15 per cent at a steady state of for continuing release at present levels. These models predict that about a 2 per cent depletion has already occurred which is below the detection limit of present technology. There is greater accuracy in the chemical kinetics involved, and additional ClO measurements have been made which are more consistent with the model. However, as a result of the increase in knowledge of the complexity of the atmosphere, the problems now under consideration are even more complicated and sophisticated than those initially dealt with.

There have been improvements in the models and in the measurements used as inputs to, and checks for, the models.

There is increasing, but still inadequate knowledge on possible effects of increased UV-radiation on ecosystems and plants, which may be the most serious impact. There is a high degree of evidence for the relationship between UV-B radiation and non-melanoma skin cancer in man, and there is some indication that there is a connection for melanoma.

RECOMMENDATION

On the basis of last year's assessment findings, the Committee had recommended : Policy makers should consider these findings and take such action as may be deemed appropriate as this time. A comprehensive environmental assessment of the impact of ozone depletion including the economic aspects of corrective and alternative actions should be considered.

Considering the present state of scientific knowledge, the Committee maintains and reinforces this recommendation. Following the second session of this Committee, the Governments of major CFM producting countries at the International Conference on Chlorofluoromethanes in Munich, 6-9 December 1978, agreed inter alia, to a significant reduction of CFM emissions. The Committee welcomes this responsible step of the Governments and proproses prompt implementation. Governments are encouraged to undertake all efforts to reach as large a reduction as seems possible. Provision of annual production figures of CFMs 11 and 12 and their major uses to UNEP were also agreed to at the Munich CH_3CCl_3 conference. It is proposed to continue this procedure and to include other relevant components e.g. CH_3CCl_3. The producing companies are requested to provide this data to the national Governments.

RECOMMENDATIONS FOR FURTHER WORK

1. Develop higher dimensional models and compare the results.
2. Extend the measurements of rate constant over the pressure and temperature range found in the stratosphere, and identify the reaction products.
3. Search for and investigate any additional reactions which may affect stratospheric chemistry.
4. Continue efforts to increase understanding of tropospheric chemistry.
5. Undertake simultaneous in situ measurements of the relative concentrations of photochemically-related compounds of the various families.
6. Maintain efforts to increase total chlorine and total fluorine measurements.
7. Undertake measurements of hydroxyl concentrations in the troposphere and stratosphere.
8. Continue to develop tropospheric monitoring to assess the lifetimes of stable species.
9. Significant further effort is needed to :
   (a) Extend UV and ozone monitoring and assess over a significant period of time, current dose-response relationships of skin cancer to UV radiation over as wide a latitude range as possible;
   (b) Develop the information to the point where reasonably reliable extrapolations can be made of the potential hazard to man from a UV radiation increase;
   (c) Increase research studies on the biological effects of UV-B on plants, animals, other organisms and ecosystems.
10. To initiate an international epidemiological study of the incidence of skin cancer, an expert panel be assembled by WHO to propose details of unified protocols and analysis of data, and to be available as an advisory body to the various countries and international organizations when data will be collected and analyzed.

11. To initiate a co-ordinated international programme relating to biological effects of UV-B on plants, animals and ecosystems.
12. Improve the total ozone monitoring by redetermination of ozone absorption coefficients, intercomparisons of Dobson spectrophotometers, add new stations optimally distributed, and maintain uninterrupted satellite total ozone measurements.
13. Establish and expand vertical ozone monitoring by development of automatic multi-wavelength ("short-Umkehr") observations, together with an expanded balloon-borne and rocket-borne ozone-sonde programme essential for providing ground truth for the satellite vertical ozone determinations.
14. Re-establish the co-operative meteorological rocket network essential for the continued development of 2-D and 3-D models.
15. Improve and expand the development of standardized, quality-controlled data sets for input to and validation of model predictions, particularly aimed at the reduction of sources of uncertainties in the measurements programme and model verification.

APPENDIX III

Principal findings of the report by the United States Academy of Sciences entitled "Stratospheric Ozone Depletion by Halocarbons, Chemistry and Transport".

There is agreement with previous reports that continued release of halocarbons into the atmosphere will result in a decrease in stratospheric ozone.

New values for some of the chemical rate coefficients have increased the predicted ozone reduction resulting from continued release of chlorofluoromethanes (CFMs).

The most probable value calculated for the eventual ozone depletion due to continued release of CFMs at the 1977 level is 16.5 percent. This value is obtained from the value of 18.6 percent calculated from the computer model allowing for possible tropospheric sinks for CFMs and for the effects on stratospheric chemistry of the CFM greenhouse effect.

There have been considerable improvements in the computer models and in the laboratory and atmospheric measurements, which have reduced the uncertainty range.

Although there are a few exceptions, the comparison between the models and measurements of substances in the present stratosphere is considered to be satisfactory within the uncertainties of the measurements. We therefore believe that the projections for ozone depletion are valid within the stated uncertainty ranges.

The uncertainties in the chemical rate coefficients, in atmospheric transport, and in the use of one-dimensional models have been combined to give an overall uncertainty range of a factor of 6 within a 95 percent confidence level.

The uncertainty range means that for the case of continued release of CFMs at the 1977 level there is 1 chance in 40 that the ozone depletion will be less than 5 percent and 1 chance in 40 that it will be greater than 28 percent.

Even allowing for the best professional judgment of the possibility that some important chemical reaction has been overlooked or that there remain large errors in the measured chemical rate coefficients, we believe that there is a 3 out of 4 chance that continued release of CFMs at the 1977 level will result in an ozone depletion that lies in the range of 9 to 24 percent.

If the rapidly increasing use of F-22 (CHF_2Cl) and methyl chloroform (CH_3CCl_3) continues unabated, the release rates and atmospheric behaviour of these compounds will require careful attention. However, since chemical reactions remove appreciable fractions of these compounds before they reach the stratosphere, substitution of F-22 for F-11 ($CFCl_3$) and F-12 (CF_2Cl_2) would be beneficial.

It is unlikely that direct measurements of the average global ozone amount would permit detection of a decrease of less than 5 percent attributable to human activity. Total cessation of CFM release at the time of detection would result in a decrease of ozone of about 7 percent some 15 years later.

APPENDIX IV

Summary of a report by the FAA (High Altitude Pollution Program).

This is the second biennal Federal Aviation Administration (FAA) report prepared in accordance with the Stratospheric Ozone Protection provisions of Public Law 95-95, the Clean Air Act Amendments of 1977.

The impact of high altitude aviation on stratospheric ozone is now believed to be an increase in total columnar ozone for flights up to 20 km (about 66,000 feet). This result has been brought about through improvements in one-dimensional chemical kinetics-transport models of the stratosphere. The improvements have consisted of including (1) odd chlorine chemistry, (2) simplified tropospheric methane oxidation ("smog") chemistry and (3) recent revisions in chemical rate constants. The cumulative effect of these improvements is that aircraft injected pollutants (primarily nitrogen oxides) generate ozone through simplified "smog" mechanisms in the upper troposphere and lower stratosphere while depleting in through the classic catalytic destruction mechanism at higher altitudes. The net result at present is a slight overall ozone increase.

The uncertainty in the present model calculations, however, is unknown.

The greatest uncertainty for the aircraft problem is the inherent inability of one-dimensional models to resolve the latitude dependence of the effects. For, while one-dimensional models allow variation of atmospheric properties (such as temperature, motion and trace species concentrations) only in the vertical dimension, both the natural ozone distribution and the aircraft injection are altitude and latitude dependent. Thus two-dimensional models, which include variations of atmospheric properties across the latitudes, are needed for reliable assessments of aircraft effects.

In addition, the following factors contributing to uncertainties in model calculations have been identified :

(1) Pressure and inverse temperature effects on bimolecular radical-radical reactions.
(2) Lack of adequate data for reaction rates in methane oxidation chemistry.
(3) Lack of atmospheric data on odd nitrogen species (e.g., nitrogen oxides) and partitioning among them for quantifying relative increases, over background levels, due to aircraft operations and for verifying model predictions.

The effort of the High Altitude Pollution Program of the Federal Aviation Administration is aimed at resolving these uncertainties so that a "consensus" method or methods can be obtained for future assessments of the impacts of high altitude aviation on the environment.

APPENDIX V

Summary and conclusions of a UK report published in 1979 by the Department of the Environment (STRAC).

INTRODUCTION

420. It has been known for some time that the chlorofluorocarbons CFC 11 and 12 ($CFCl_3$ and CF_2Cl_2) which find extensive use as aerosol propellants, refrigerants and from blowing agents accumulate in the lower atmosphere (the troposphere) where they have a long lifetime. In 1974 Molina and Rowland pointed out that these compounds would eventually be transported upwards into the stratosphere where they would be decomposed by far ultraviolet light to yield free chlorine atoms. According to their calculations these chlorine atoms would destroy significant quantities of stratospheric ozone before being converted to HCl which is removed from the troposphere by rain.

421. Making the assumption that there were no other significant removal processes for CFC 11 and 12 and using a simple one-dimensional model of the atmosphere, Rowland and Molina (1975) deduced that continued release of these compounds at the 1973 production rates would eventually lead to a globally-averaged total ozone reduction of about 10%. This would take between 50 and 70 years to be reached since, when fully mixed, only 10-20% of the atmospheric burden of the CFCs could be in the stratosphere at a given time and it would take several years for them to pass through their stratospheric decomposition cycle. The high variability in ozone amounts makes it difficult to detect slow changes in total ozone. Because of the long time predicted to reach maximum reduction, it has been argued that uses of CFCs must be restricted without waiting for ozone depletion to be detected.

422. The hypothesis of possible ozone reduction was supported by other assessments, such as Pollution Paper No 5, in which the same basic assumptions were made but with some differences in the parameters used in the model calculations. However, as was pointed out in the NAS Report (1976) and the subsequent NASA re-evaluation (1977), the fact that a number of independent workers who used these same assumptions calculate eventual ozone depletions in the same range (usually 5-20%) does not prove that the true figure would lie in this range. Laboratory data on the rates of chemical and photochemical processes which are used in all the modelling calculations may prove incorrect, important processes may have been neglected and the simplifications used to model transport processes may be injustified.

423. The aim of this report has been to make a more complete assessment of current understanding of the problems involved and the success or otherwise of obtaining realistic estimates from models of the possible reductions of stratospheric ozone by halocarbons. This includes, in particular, a detailed consideration of many of the assumptions in such studies. The past four years have seen intense research activity which has confirmed the basic postulate that CFCs are transported upwards to the stratosphere where they are broken down to yield, according to laboratory studies, active chlorine species which would catalytically decompose ozone. There is evidence consistent with the view that chlorine is removed from the stratosphere as HCl which is subsequently rained out. In addition the models have successfully simulated some of the major features of the distribution of ozone in the unperturbed atmosphere.

424. However, although considerable progress has been made and the continuing research has narrowed uncertainties in some areas it has indicated important gaps in our understanding of the problem in its entirely. The factors affecting the reability of current quantitative predictions of ozone perturbations are considered in the following sections.

Sources of chlorine in the atmosphere

425. The Manufacturing Chemists Association has published details of the global production of CFC 11 and 12 between 1931 and 1977 with a claimed accuracy of ± 5%, and has estimated the fraction employed in each of the major end uses. The assumption of continuing release at either the 1975 or the average 1973-1976 production rate is the usual basis for predicting possible ozone reductions at steady state. Assessments of the delay before releases to the atmosphere for each end use are uncertain and vary from about six months for aerosol propellants to 12 (± 4) years for hermetically sealed refrigerators. The release estimates are significant because, when combined with measurements of the global atmospheric CFC burden, they can lead to evaluation of sink strengths.

426. Other compounds such as methyl chloride, methyl chloroform and carbon tetrachloride make a major contribution to the current chlorine budget in the stratosphere. Methyl chloride is of natural origin, methyl chloroform is man made and the respective contributions to carbon tetrachloride are uncertain.

Tropospheric removal processes

427. Processes that remove CFCs from the troposphere have considerable potential importance. The great bulk of the CFCs reside there and a relatively slow tropospheric sink would compete with a rapid stratospheric sink. The NAS report (1976) concluded that no known tropospheric removal process could compete with the stratospheric removal. Since then decomposition of CFCs absorbed on desert sand has been demonstrated in the laboratory. The evidence as to whether or not a significant similar sink exists in the atmosphere is not conclusive and its quantitative status will be difficult to establish (paragraphs 346-348).

428. The atmospheric concentrations of CFCs are being monitored in an attempt to determine their atmospheric lifetime and whether or not there is any significant removal process in the troposphere (paragraph 59).

429. Our knowledge of the removal of other atmospheric halocarbons from the troposphere is also unsatisfactory. Possible mechanisms are deposition on the earth's surface, removal by the oceans, photolysis by solar radiation and attack by OH radicals. The last is potentially important for those halocarbons with C-H or C-C linkages, some of which have been proposed as substitutes for CFCs 11 and 12, e.g. CFC 21 ($CHFCl_2$) and CFC 22 (CHF_2Cl) but cannot be accurately assessed until data on tropospheric OH concentrations become better established.

430. The AERE Harwell 2-D tropospheric model estimates shorter tropospheric lifetimes for CFCs 21 and 22 compared with those of CFCs 11 and 12. Thus these compounds appear to offer some potential as substitutes but may be unsatisfactory for other reasons (paragraphs 391-395).

STRATOSPHERIC CHEMISTRY

431. Our knowledge of the rates of about 100 reactions so far identified as influencing ozone concentrations has improved steadily over the last few years as a result of extensive laboratory measurements. The uncertainty in model predictions arising from errors in these chemical and photochemical data is relatively easy to assess. It was estimated by NASA (1977) as a factor of two upwards and three downwards. In future the error band should narrow steadily but no dramatic improvement is likely. In contrast the uncertainty from the different assumptions made in simulating the transport processes and in the modelling procedures cannot yet be quantified, nor can any firm statement be made regarding the uncertainties arising from possible deficiencies in our knowledge of the detailed chemistry and photochemistry.

432. There are discrepancies between the calculated results and the measurements at present available. These include examples taken from the discussions of the major families of species : O_x, HO_x, NO_x and ClO_x.

(i) For chemical and/or dynamical reasons the total ozone column and the vertical distribution of ozone have not yet been accurately calculated in the models (paragraphs 176-188). Furthermore, the models considerably underestimate the reported $O(^3P)$ concentration in the middle stratosphere (paragraphs 189-191) due partially to the neglect of molecular scattering in the STRAC models. There are also reservations about the calculated rate of $O(^1D)$ production from the photolysis of ozone (paragraph 20).

(ii) Measured OH and HO_2 concentrations imply a greater odd-hydrogen production rate than can be sustained by the current photochemical schemes (paragraphs 194-200). Several recent observations of water vapour indicate (though with considerable spread) appreciably higher concentrations in the lower and middle stratosphere than calculated in the models (paragraphs 192-193). This might explain the higher observed OH and HO_2 concentrations, but would leave open the question of the possible origin of this additional water vapour and the effect the higher OH and HO_2 concentrations may have on the calculated distributions of other trace gases in the stratosphere.

(iii) Laboratory studies of the combination reactions of NO_2 with ClO, NO_3 and HO_2 have suggested that their products (chlorine nitrate, nitrogen pentoxide and peroxynitric acid) should be formed in detectable quantities. There has been a tentative identification of $ClONO_2$ but there is so far no convincing evidence for the presence of N_2O_5 and HO_2NO_2 (paragraphs 228-232).

(iv) A comparison of observation with model calculation for Cl, HCl, ClO and HF reveals discrepancies whose origins may lie in the inadequency of present understanding of stratospheric chemistry (paragraphs 213-221 and 273-277). This inadequacy may well cause significant errors in the estimation of ozone reductions in the upper stratosphere from the chlorine species liberated in the degradations of CFCs.

433. There are many possible reasons for the above discrepancies. Although a considerable body of reliable photochemical and kinetic data has been established under appropriate laboratory conditions it is possible that not all the processes appropriate to the atmosphere have been identified and investigated.

TRANSPORT AND MODELLING

434. There are many inadequacies in the representation of transport and the procedures used in the current models. Some are listed below.

(i) Errors are introduced by the poorly based flux-gradient (K) concept and the application of the same values of K of all species without verification (paragraphs 255-258).

(ii) The averaging procedures used to obtain the chemical kinetic terms in the models do not take account of the covariance of reactants associated with the inhomogeneity of the stratosphere. As a result the effective rate of reactions in the model calculations contain errors whose magnitude has not been established (paragraphs 244-254).

(iii) The feedback mechanisms inherent in the interactions between the stratospheric composition, radiation, temperature and motion field are not adequately represented.

(iv) Long-term atmospheric variability, for example, sudden warmings, quasi-biennal oscillations, interannual changes, is neglected.

(v) The solar radiation transfer may be inadequately formulated especially the effects of scattering and the description of the penetration of Solar radiation through the atmosphere.

435. The modelling techniques adopted in the UK and abroad all involve major assumptions (paragraph 145). It is not possible to assign numerical values to the associated uncertainties and this limits the reliance that can be placed on current models for predictive purposes.

MODEL PREDICTIONS

436. Although often used, a single average figure for ozone reduction is an over-simplification. For example, the Oxford University and Meteorological Office models show that significant variations of the ozone reduction with latitude and season are to be anticipated. The maximum reduction occurs in late winter at high latitude and the minimum at low latitudes; these may differ by a factor of about four.

437. With the above reservations, the calculated reduction to date from CFCs 11 and 12 is around 1%, being 1.3%, 1.6% and 0.7% in the three UK models. If releases from all sources were stopped immediately, the reduction is predicted to reach a peak of around 2% about ten years from now. Delay of similar action until 1983, for instance, would result in a further reduction of 0.5% to a peak of about 2.5%.

438. Should global emissions continue indefinitely at about the 1975 rate a steady state would be reached with ozone being reduced by an amount estimated as 13% in the AERE Harwell 1-D model, 11% in the Meteorological Office 3-column model and 16% in the Oxford University 2-D model. The predicted reduction would reach half of its peak figure

after about fifty years. Experiments conducted with the AERE Harwell 1-D model show that ozone reductions are roughly proportionnal to CFC emissions. If, for example, global emissions continued indefinitely at half the 1975 level, the predicted steady state depletion would be approximately halved.

439. It is not possible to assign uncertainty ranges to these estimates. To give some idea of the difficulties involved we note that the AERE Harwell 1-D model gives values in the range 6-22% simply by using different published eddy diffusion profiles. The Meteorological Office 3-column model shows comparable changes when the two-dimensional eddy diffusion coefficients are similary changed. The Oxford University 2-D model shows considerable sensivity to the extrapolation procedure used to estimate the steady state ozone depletion with a range of 8% to 16%. It also shows sensivity to the inclusion of coupling between ozone and the temperature and wind field (paragraphs 331-339).

440. It is not realistic to consider global ozone reductions in terms of the effects of CFCs 11 and 12 releases . Many other factors may increase or decrease the global ozone amount, including : other halocarbons; CO, NO_x and CO_2 from combustion; N_2O from fertiliser application and changes in land use; bromine compounds used as fumigants and fire retardants; and natural perturbing effects such as the solar cycle, lightning effects, cosmic ray produced NO_x and changes in the global circulation. It is their combined effect which will determine what will actually happen to the ozone layer.

441. For instance, it has been calculated that stratospheric temperature decreases resultant upon increasing CO_2 in the atmosphere due to burning fossil fuel and deforestation may significantly affect stratospheric ozone. Generally decreases in stratospheric temperatures are believed to be linked to an increase in the ozone amount. When radiative-convective effects due to projected CO_2 increase are included

together with CFCs in the Meteorological Office 1-D model, decrease of about a factor of two in the ozone reduction estimate is found compared with that deduced when CO_2 is held constant.

442. The AERE Harwell 1D model has been used to estimate the continued effect of CFCs and N_2O soil emissions following nitrogen fertiliser application at the rate of 200 million tons per year. The model predicts that the ozone depletion increases by a factor of about 1.1-1.2 compared with that deduced when N_2O is held constant.

443. The effect on ozone of man-made chlorine containing compounds other than CFCs 11 and 12 has also been calculated. Assuming production of these compounds at 1977 levels, their effect is to increase the predicted steady state ozone reduction. The combined ozone reduction due to non-natural chlorine compounds is about one-third of that due to CFC 11 and 12 alone.

444. The uncertainties in the model calculations of ozone reductions due to CFCs apply with equal force to the calculations of the effects of these factors.

445. Ozone reduction estimates are not simply related to the possible increased penetration of solar ultraviolet radiation to the surface. The latitudinal and seasonal variations in the total ozone reduction shown by the two-dimensional models employed here imply that the simple working relationship used formerly (see for example in Pollution Paper No 5), that a 1% decrease in total ozone results in a 2% increase in UV-B at the ground may no longer be appropriate (Chapter 8).

ATMOSPHERIC MEASUREMENTS

446. There is a requirement to obtain sufficient measurements to check model result for species other than ozone and to search for

possible undiscovered substances. The measurements required include diurnal, seasonal, latitudinal and sporadic changes of substances at present measured and the simultaneous measurements of many other species for which there are, so far, few data. To be effective, the latter should be obtained from the same volume of atmosphere (e.g. because local grab samples are evidently not directly comparable with longpath averages). In order to assess the significance of the data, it is essential to make use of both long-path and local methods, so that averaged data can be compared with local variations. To determine temporal trends, observations must be repeated at the same locations, or at least the same regions, since geographical variations are certain to be significant.

447. The present position regarding measurement techniques is unsatisfactory to the extent of limiting possible progress. There are a number of incompatibilities between atmospheric measurements and model predictions. This position is unsatisfactory and there is a need to establish a sound measurement base so that we can determine the cause of the discrepancies. Such a measurement base should have a good temporal and spatial coverage and be verified by independent techniques.

448. As well as expanding the measurements of short lived species necessary to validate the models and establishing programmes determine long term trends, there is a need to build up a continuing series of measurements of natural and manmade source species e.g. N_2O, CH_4, CO, NO_x, CO_2, CH_3Cl as well as the CFCs and CCl_4. Such measurements are under way and research projects initiated over the past two to three years now reaching fruition. Significant results from these programmes may become available in the next few years.

OZONE MEASUREMENTS

449. The model calculations indicate that it will be another 5 to 15 years before the CFC releases at present rates will lead to an ozone reduction of 2% and about 40 years before a 6% reduction might be caused. These should be compared with estimates of detectability of global ozone changes (paragraphs 140-142).

If these two sets of estimates are correct (viz ozone depletions and detectable changes) there could be no firm indication of ozone reductions within the next decade. It is, however, clear that if the larger ozone reductions that have been predicted by some other models were to occur, they would be more reality detected. It should also be noted that the small extra decreases in ozone amounts likely to result from continuing current releases of CFCs for a few more years could not be resolved from studies of the global ozone record. Even if detected, it would be impossible to ascribe a small decrease in ozone amounts to CFC release alone. The historical ozone record shows a considerable natural variability and the trend, if there has been any over the past two decades, has been of increasing ozone.

GREENHOUSE EFFECTS

450. At the time of writing no further work has been published on the socalled greenhouse effect caused by the strong infrared absorption bands of CFC 11 and 12 in the atmospheric window region (8-14 μm wavelength). It seems likely that this effect on the climate will be small compared to that which might be caused by atmospheric CO_2 increases over the time scales of interest in this report.

CONCLUSIONS

451. Current predictions of the theoretical studies by STRAC modelling groups predict reductions in stratospheric ozone amounts of the order of 11-16% in steady state for release of CFCs at present levels. On the basis of these calculations, CFCs would have reduced stratospheric ozone by about 0.7-1.6% already. If all releases were discontinued immediately the reduction is predicted to reach a peak of about 2% 5-15 years from now. If releases continued for a short time, until 1982, this peak figure would increase by 0.5% to 2.5%. Any uncertainty analysis would be incomplete as it would encompass only quantifiable sources of uncertainty.

452. The results concur with other studies that predicted reductions in the total ozone amount are greater than those estimated in 1975 at the time of the preparation of Pollution Paper No 5. Whether the statements realistically describe what is likely to happen in the atmosphere depends on the validity of the calculations and also their coverage including the simultaneous effects of other atmospheric pollutants. Basic scientific understanding, although progressing rapidly, is still inadequate in many respects.

453. Understanding of the stratosphere has been greatly accelerated during the last decade, largely as a result of environmental concerns. It is important that this effort should continue, but it is not possible to predict how rapidly this will give definitive answer to specific environmental problems.

454. In view of the great natural variability in the stratospheric ozone amount and the many phenomena that can effect it, there seems little prospect of detecting the ozone changes of the magnitude predicted by the models within the next decade, though, if the changes were greater then it should be possible to detect them earlier. There would,

however, be no certainty that any change detected was due to CFCs. If it were, the time lag in the CFC-ozone system is such that, after curtailing CFC release, the predicted ozone depletion would reach a maximum 5-15 years later and then decrease over a period of 50-100 years. The maximum ozone depletion would be greater than the minimum that could be detected by a factor of 1.2-3 or less.

European Communities - Commission

EUR 7067 - Analysis of recent reports on the effect of chlorofluorocarbons on atmospheric ozone

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It has been known for the past few years that chlorofluorocarbons (CFCs) have been accumulating in the atmosphere, as a result of their releases in uses such as aerosol propellants, refrigerants and foam-blowing agents. According to theories developped in US and Europe, the CFCs slowly migrate into the stratosphere and destroy the ozone layer which protects us against UV-radiation. Many scientific reports have been written on this subject, but the opinions sharply differ in certain cases. The aim of this analysis is to compare conclusions reached by the various reports in order to take stock of the certainties and uncertainties surrounding the problem of ozone. An attempt is then made to put forward a number of projects to be carried out within the next five years.