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Summary of recent research results  
relevant to the World Plan of Action  
on the Ozone Layer

Contribution

by the

United Kingdom of Great Britain  
and Northern Ireland

A continuing programme of research is maintained in the UK into the behaviour of the stratospheric ozone layer, the perturbing effects thereon of various gaseous species and consequential changes in ultra-violet radiation at the earth's surface. The programme includes atmospheric measurements, chemical kinetic studies and numerical modelling. Overall assessment of the ozone depletion problem is also carried out.

1. Atmospheric Measurements

(i) Ground-based measurements

Routine total column ozone measurements have continued from stations operated by the METEOROLOGICAL OFFICE (MO) at Bracknell, Lerwick, St Helena and Mahé, Seychelles. These are published by the World Ozone Data Centre, Downsview, Ontario.

The MO Dobson spectrophotometer on loan to the Singapore Meteorological Service was visited and repaired in September 1980 and is now returning regular observations again.

A MO technician visited the instrument on St Helena for overhaul and recalibration against the MO standard instrument in December 1980. An additional instrument ordered by the British Antarctic Survey has been received at Bracknell, calibrated and compared with our standard instrument.

The instrument from Magny les Hameaux visited Bracknell in June 1981 for checking and adjustment before being taken to the Observatoire de Haute Provence for a comparison of different ozone measuring techniques in July.

Arrangements have been made for the continuation of ozone measurements from the Seychelles after the withdrawal of British Meteorological Office staff in August 1981.

The BRITISH ANTARCTIC SURVEY (BAS) have continued processing the Dobson spectrophotometer observations made at Grytviken, 54°S, Faraday 65°S and Halley, 76°S, sending the results to the world centre in Toronto.

This coming Antarctic season, BAS plan to launch approximately 70 ozone sondes for NASA. This program is to be conducted from Faraday at

times when suitable instrumented satellites are flying overhead.

HARWELL have continued their long-term measurements of CFC11 and  $\text{CCl}_4$ . The data set now covers the period January 1975 to June 1981. Measurements of methylchloroform ( $\text{CH}_3\text{CCl}_3$ ) have recently been added to the programme. The data indicate that the rate of growth of CFC11 in the atmosphere has slowed down noticeably over the past two years.

The present background level at Harwell is estimated to be about 185-190 ppt (parts per  $10^{12}$ ). The  $\text{CCl}_4$  data can be accounted for on the basis of industrial sources only, there being no evidence for a natural contribution.

The long-term measurement of  $\text{N}_2\text{O}$  on a regular basis has also continued at HARWELL.

In a cooperative programme between HARWELL and BAS air samples are being obtained on a latitudinal transect between  $50^\circ\text{N}$  and  $50^\circ\text{S}$  using an Antarctic supply vessel. Samples are being analysed for a range of halocarbons and hydrocarbons.

(ii) Aircraft-based measurements

The air samples obtained earlier by HARWELL using a commercial Lear-jet have since been analysed. They have established a concentration of 50 ppt for CFC 22 ( $\text{CHF}_2\text{Cl}$ ) in the upper troposphere which is unaffected by local sources<sup>(1)</sup>. This is substantially greater than predicted from release estimates ( 35 ppt) and suggests these should be re-examined.

The measurement programme on long-lived tropospheric precursor molecules carried out by the METEOROLOGICAL OFFICE is continuing using an airborne gas chromatograph on the Cl30 Hercules aircraft of the Meteorological Research Flight.

(iii) Balloon-borne measurements

On 25th September 1980 a balloon probe was launched from Aire sur L'Adour in Southern France (latitude  $44^\circ\text{N}$ ) to collect large air samples at a number of altitudes from 33 km down to 10 km. The flight, funded

by the UK Dept. of the Environment, was carried out in cooperation with the Max-Planck-Institute for Aeronomy at Lindau, FGR who provided the air sampling equipment. About 20 litres of air (at STP) was collected at 7 altitudes, 9.9, 14.4, 20.1, 23.3, 25.9, 28.8 and 33.3 km by cryogenic collection using liquid neon. The collected air was subsequently transferred to 1.6 litre stainless steel bottles proven to be contamination-free. The samples were later analysed at HARWELL using gas chromatography and gas chromatography/mass spectrometry (GC/MS)<sup>(2)</sup>.

A total of 26 different chemical species were analysed which included vertical concentration profiles for CH<sub>4</sub> and H<sub>2</sub>O, and simultaneous measurement of a large variety of chlorine containing molecules. Amongst these were CFC1<sub>3</sub> (CFC 11), CF<sub>2</sub>Cl<sub>2</sub> (CFC 12), CF<sub>3</sub>Cl (CFC 13), CF<sub>4</sub> (CFC 14), C<sub>2</sub>F<sub>3</sub>Cl<sub>3</sub> (CFC 113), C<sub>2</sub>F<sub>4</sub>Cl<sub>2</sub> (CFC 114), C<sub>2</sub>F<sub>5</sub>Cl (CFC 115), C<sub>2</sub>F<sub>6</sub> (CFC 116), CF<sub>3</sub>Br (CFC 13B1), CFH<sub>2</sub>Cl (CFC 22), CH<sub>3</sub>Cl, CHCl<sub>3</sub>, CCl<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>Cl<sub>2</sub> and CH<sub>3</sub>CCl<sub>3</sub>. The data for some of these compounds are shown in Table 1. The profiles for CFCs 13, 14, 22, 113, 114, 115 and 116 and for CH<sub>3</sub>CCl<sub>3</sub> are the first reported and will be of particular value for validation of two-dimensional stratospheric models.

Table 1

Vertical concentration profiles for CFCs, CH<sub>3</sub>Cl, CH<sub>3</sub>CCl<sub>3</sub> and CCl<sub>4</sub>  
Balloon flight September 25th 1980 at Lat. 44°N

Altitude	CH <sub>3</sub> Cl ppt	CFC11 ppt	CFC12 ppt	CFC13 ppt	CFC113 ppt	CFC114 ppt	CFC115 ppt	CH <sub>3</sub> CCl <sub>3</sub> ppt	CCl <sub>4</sub> ppt
9.9 km	(477)	(144)	(241)	(5.1)	C	(7.0)	(4.2)	(78.2)	abs
14.4 km	422	178	289	3.9	10.4	8.5	3.1	44.7	abs
20.1 km	183	90.3	189	3.52	5.1	6.0	2.3	17.2	abs
23.3 km	119	14.7	129	2.99	2.73	5.4	1.7	1.04	abs
25.9 km	44	0.28	39.6	2.65	1.0	3.8	1.36		abs
28.8 km	34	0.09	20.8	2.45	0.6	4.4	1.26		abs
33.3 km	54.5	N.D.	8.96	2.26	C	2.7	1.0		abs

C = contaminated

N.D. = not detected

abs - absent

Organic compounds which react with hydroxyl radical formed a further group of compounds analysed. Apart from chlorocarbons this included ethane, ethylene, benzene, toluene and vinylidene chloride ( $C_2H_2Cl_2$ ).

The stratospheric programme at the NATIONAL PHYSICAL LABORATORY (NPL) is directed towards remote near-simultaneous measurement of the concentration profiles of inter-related stratospheric gases believed to be important to the problem of ozone depletion. A balloon-borne, mid-infrared cooled grating spectrometer is used which can measure the emission spectra of stratospheric species throughout a daily cycle<sup>(3)</sup>. This instrument employs the conventional limb-scanning technique to derive concentration profiles as a function of altitude. In its present form the spectrometer is directed towards measurements of the NOx family, though several other species are measured such as ozone, water vapour, CFCs 11 and 12 and methane. The experiment is mounted on a stabilized gondola platform developed at NPL. Azimuth stabilization is achieved by sensing the gondola position with respect to the earth's magnetic field and servo controlling the gondola rotation. Stabilization in elevation is achieved by sensing the gondola swing away from vertical and correcting the angle of the limb-scan mirror accordingly. The instrument covers the spectral region 5 to 16  $\mu m$  and has a resolution approaching  $0.1 \text{ cm}^{-1}$ ; sensitivity is high since the three copper doped germanium detectors are liquid helium cooled and the spectrometer and input window are at liquid nitrogen temperature.

In parallel with the major NOx programme has been the development of a simple, light-weight filter radiometer for the measurement of water vapour in the far-infrared. This device can be launched by two standard radiosonde balloons or lends itself to "pick-a-back" operation with larger payloads.

During the Spring of 1981 the group co-ordinated and assembled a major collaborative payload onto the NPL gondola. The combined experiment was named NOBLE (Nitrogen Oxides Balance Experiment) and consisted of:

Cooled grating spectrometer	(NPL)
Water vapour radiometer	(NPL)
Pressure modulated radiometer	(OXFORD/JPL)

Ozone sonde	(NASA, HOUSTON)
Two solar flux experiments	(NASA, GODDARD)

The experiments produced much useful data during the 16½ hour flight from Palestine, Texas. The float altitude reached was 38 km.

Also during the Spring of 1981 a campaign took place to make near simultaneous intercomparison flights of instruments measuring water vapour. Seven groups participated, five balloons were launched and a U - 2 flight took place. The NPL water vapour radiometer flew on three of the balloon payloads and produced good data.

The results of a balloon flight carried out by the Dept of Atmospheric Physics, OXFORD UNIVERSITY, in September 1978 at Palestine Texas have now been published<sup>(4)</sup>. The Oxford balloon-borne pressure modulation radiometer was used to measure simultaneous altitude profiles of NO and NO<sub>2</sub> from mid-morning till sunset. An improved detector for the NO channel, a better procedure for interpolating between calibration points, and an improved error analysis yielded results of higher quality than from previous flights. During the day the NO<sub>2</sub> column above 24 km increased by  $0.42 \pm 0.28$  to  $1.86 \pm 0.24 \times 10^{15}$  molecule cm<sup>-2</sup>; the NO column above 30 km reached a maximum of  $5.2 \pm 1.3 \times 10^{15}$  molecule cm<sup>-2</sup> soon after noon, falling  $1.58 \pm 0.77 \times 10^{15}$  molecule cm<sup>-2</sup> by an hour before sunset. A detailed comparison of measured NO and NO<sub>2</sub> concentrations with those predicted by the OXFORD two-dimensional diurnal model shows disagreement in their absolute values but close agreement in their day-time changes. The model predicts a fall in NO during the afternoon, as observed.

The Remote Sounding Group at the RUTHERFORD and APPLETON Laboratories (RAL) is constructing a balloon-borne microwave radiometer system for upper tropospheric and stratospheric measurements of trace species

(iv) Satellite measurements

The Stratospheric and Mesospheric Sounder (SAMS) designed and constructed by the Atmospheric Physics Group, OXFORD UNIVERSITY<sup>(5,6)</sup> is an infra-red radiometer on the NIMBUS 7 experimental meteorological satellite which was launched on October 24th 1978. It has completed two years of successful operation and preparations are now being made to go into full-scale production of archivable data soon to be available

on magnetic tape. Data acquisition is expected to continue throughout the Middle Atmospheric Programme (ie until 1985) and possibly until the launch of an advanced version (ISAMS) on the first Upper Atmospheric Research Satellite in 1987. The data reduction and inversion algorithms will also be subject to continuous improvement as time goes on and so occasional reprocessing of the data obtained since launch, with a consequent revision of the inferred geophysical parameters, is anticipated.

Like other equipment on NIMBUS, SAMS is an experimental, not an operational, instrument. It was designed to measure temperature and chemical composition over a wide range of altitudes, using the pressure modulation technique to give high selectivity and sensitivity for the constituents of interest. The molecular species and spectral bands selected for SAMS are summarized in Table 2, which also lists the height ranges over which measurements typically can be made in each case.

TABLE 2. S.A.M.S. RADIOMETRIC CHANNELS

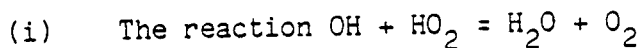
field of view	channel	constituent (gas in modulator)	mean cell pressures	spectral band	derived quantities and altitude range
			mbar	$\mu\text{m}$	
A	A1	CO <sub>2</sub>	17, 2.4, 0.89, 0.25	15	kinetic temperature 15-80 km; attitude $\nu_2$ vibrational temperature 80-100 km
	A2				
	A3	CO	14.8, 4.5	4-5	distribution 15-60 km
	A4	NO	45, 20		
B	B1	H <sub>2</sub> O	10, 4.5, 0.8, 0.5	2.7	distribution 80-100 km
	B2			25-100	distribution 15-100 km
C	C1	CO <sub>2</sub>	36, 11.2, 3.25, 0.87	15	kinetic temperature 15-80 km; attitude distribution 15-80 km
	C2	N <sub>2</sub> O	24.4, 7.15	7.7	
	C3	CH <sub>4</sub>	47.8, 22.5		

The Remote Sounding Group at RAL is assessing the H<sub>2</sub>O and NO<sub>2</sub> + HNO<sub>3</sub> data from the LIMS infra-red experiment, also carried on NIMBUS 7, to determine global composition distributions and budgets.

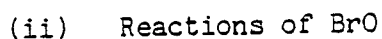
## 2. Laboratory Measurements and Chemical Kinetics

Laboratory spectroscopy of H<sub>2</sub>O, O<sub>3</sub> and other trace molecules is being carried out by RAL to provide spectral data needed for remote sensing of the stratosphere.

At HARWELL the technique of molecular modulation spectroscopy (MMS) has been upgraded by improved data handling facilities and its versatility will shortly be extended by the use of an infra-red diode laser to provide diagnostic capability in the infra-red region to supplement the existing visible and ultra-violet detection systems. The MMS continues to be used to obtain kinetic and mechanistic information.

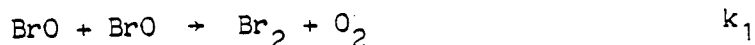


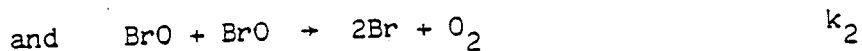
The kinetics of this reaction have been the subject of numerous studies over a number of years. Measurement of the rate coefficient is experimentally difficult and there remains substantial disagreement between the apparent rate coefficient obtained at high pressure (1 atm) and the value from low pressure flow systems. This reaction plays a key role in the photochemically driven odd-hydrogen chemistry at all altitudes of the atmosphere, since it leads to a net removal of OH, the most important species promoting the oxidative degradation of many trace gases. The use of the upgraded multichannel detection system for the molecular modulation spectrometer in which the fully time-resolved waveform for absorption is recorded, has greatly facilitated the extraction of kinetic information from this kinetically complex reaction<sup>(7)</sup>. A value of  $(9.9 \pm 2.5) \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  at 308°K and 1 atmosphere pressure was obtained in excellent agreement with recent values near  $1.0 \times 10^{-10} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  obtained elsewhere from investigations conducted at high pressure using steady-state photolysis, flash-photolysis and pulse-radiolysis. It is however significantly higher than the range 2 to 5  $\times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  based on a number of low pressure discharge flow experiments.



The absorption spectrum of BrO was determined<sup>(8)</sup> from measurement of the in-phase components of modulated absorption in the photolysis of  $\text{Br}_2\text{-O}_3\text{-O}_2$  mixtures. The observed spectrum consisted of 17 bands in a progression from 361 nm to 297.6 nm. Using standard data for the solar flux near the ground the photolysis rate of BrO was calculated to be  $J(\text{BrO}) = 3.4 \times 10^{-2} \text{ s}^{-1}$  for a solar zenith angle of 30°.

The kinetics of the reactions





were investigated<sup>(8)</sup> by photolysing  $\text{Br}_2$  in the presence of  $\text{O}_3$ . From measurements using the multichannel detector to monitor  $\text{BrO}$  the rate coefficient  $k_1$  was determined as  $(6.6 \pm 2.0) \times 10^{-13} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  over the temperature range 278-348°K and by measuring the quantum yield for the  $\text{Br}_2$  photo-sensitised decomposition of  $\text{O}_3$  at 303°K, the value of  $k_2$  was estimated as  $3.5 \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ .

The reaction of  $\text{BrO}$  with  $\text{HO}_2$  was studied by simultaneous production of these radicals in the photolysis of  $\text{O}_3\text{-O}_2\text{-Br}_2\text{-H}_2$  mixtures. Concentration-time data for  $\text{BrO}$  and  $\text{HO}_2$  monitored in separate experiments at different monitoring wavelengths and under conditions where the photochemical production rates for the two species were nearly equal, were fitted by computer simulation to obtain an estimate of  $4.3 \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$  for the reaction  $\text{BrO} + \text{HO}_2 \rightarrow \text{HOBr} + \text{O}_2$ .

Bromine is a potentially powerful catalyst for destruction of ozone via chain reactions involving  $\text{Br}$  and  $\text{BrO}$ . Although the photolysis rate for  $\text{BrO}$  determined in this study constitutes a significant reaction of  $\text{BrO}$  in the stratosphere, its effect on the catalytic role of bromine species in  $\text{O}_3$  depletion will probably be small since it does not give rise to loss of active  $\text{Br}$ . However the reaction of  $\text{BrO}$  with  $\text{HO}_2$  is postulated to yield  $\text{HOBr}$  which may act as a temporary reservoir species. This would tend to reduce the concentration of active  $\text{Br}$ .

### 3. Numerical Modelling

At the METEOROLOGICAL OFFICE examination of the correlations between water vapour and ozone in a stratosphere-troposphere general circulation model has been carried out, and the effects on the global distribution of  $\text{OH}$  calculated<sup>(9)</sup>.

Detailed calculations of the diurnal, seasonal and semiannual variations of ozone and temperature have been made in the stratosphere with a radiative-photochemical column model at a range of latitudes, and the data compared with satellite observations. A preliminary account of this work is included in the collected extended abstracts of the 1980 Quadrennial Ozone Symposium, held at Boulder.

Also at the METEOROLOGICAL OFFICE there is work on three dimensional models with emphasis on dynamics though the interactions

and transport of minor constituents are also included.

The stratosphere-mesosphere model used as the basis for these studies has as its lower boundary condition specified geopotential heights in the tropopause region, in order to parallel the forcing provided by tropospheric planetary waves without the complexity of a tropospheric general circulation model. This model structure also conveniently allows use of the model for idealised experiments and studies of specific observed events. Comparisons of circulation with observations from the Meteorological Office's Stratospheric Sounding Units (SSUs) on board NOAA operational satellites is used both for atmospheric studies and guidance in model development. Some of this work has recently helped in elucidation of the mechanism of the wavenumber two sudden warming of February 1979. It was demonstrated in a set of model experiments that the occurrence of warming depended upon both the non-climatological circulation prior to the warming and the phase speed of the forcing wave in the troposphere. Because acceleration of the circulation and transport are intimately linked, results such as these have also implications for the fuller understanding of the behaviour of trace species.

A version of this model is also being used to study the evolution of an ozone-like tracer in the presence of a transient planetary wave disturbance. Diagnostic techniques are being developed to clarify the contributions of wave transience and induced chemical interaction to the net flux of minor constituents. Recent effort has been mainly directed towards simulating the observed circulation well enough to allow realistic description of the propagation and effects of wave disturbances. Test integrations have been able to reproduce the maintenance of the polar night jet quite well using calculated radiative forcing and observed planetary wave characteristics. These should provide a reasonably quantitative basis for further study of budgets of dynamical variables and idealised tracers.

An advanced treatment of transport processes is being attempted with the OXFORD UNIVERSITY 2D model. In this treatment the well-known cancellation between the mean and eddy transports is modelled explicitly. The net mean transport is accomplished by the diabatic circulation. Eddy contributions to net transport, which depend on the chemical lifetime of the particular constituents are also calculated from a

description of planetary wave propagation. Preliminary calculations are encouraging producing, for instance, the major features of the ozone distribution.

The HARWELL 1-D model has been continuously updated during the past year to reflect the changes in stratospheric chemistry resulting from new laboratory studies and chemical kinetic rate evaluations. Since the previous UNEP evaluation in 1980 new data for the reactions  $\text{OH} + \text{HNO}_4$ ,  $\text{OH} + \text{HO}_2$  and  $\text{ClO} + \text{NO}_2$ , together with numerous minor changes to the  $\text{O}(^1\text{D})$  rate coefficients,  $\text{NO} + \text{O}_3$  etc have been incorporated. Using the latest evaluations from the combined NASA-CODATA chemical kinetics panels, a total column ozone depletion of 6.7% is currently calculated at steady state for releases of CFC 11 and 12 continued indefinitely at their 1976 rates. This value should be compared with a figure of 10.7% calculated at the time of the 1980 meeting of the CCOL and is about one third of the prediction made in 1979 when estimates of future ozone depletion reached a peak.

The fluctuations in predicted ozone column depletion resulting from 1D model calculations carried out at various times since 1976 using on each occasion the best currently available chemical kinetics data are shown in Fig. 1. The results of some calculations by other groups are also included.

At the present time total column ozone depletion is estimated to have reached 0.65%. At 40 km, where the effect of CFC 11 and 12 are expected to be larger relatively, the depletion is estimated to be 2.5% currently and to reach 25% at steady state. If CFC 11 and 12 production rates are held constant at their 1979 levels (which were lower than in 1976) then the total column ozone depletion is not expected to reach 2% until about 2000 AD.

At present validation of the models used to predict ozone depletion can only be carried out by comparison of model and observed values for the present day levels of a number of trace species in the stratosphere. The HARWELL 1-D model has been used<sup>(10)</sup> to examine the adequacy of currently available atmospheric data for this purpose. It was shown that different model formulations which result in predicted ozone depletions differing by a factor 2 result in changes in the calculated present day levels of trace species which are well within the current

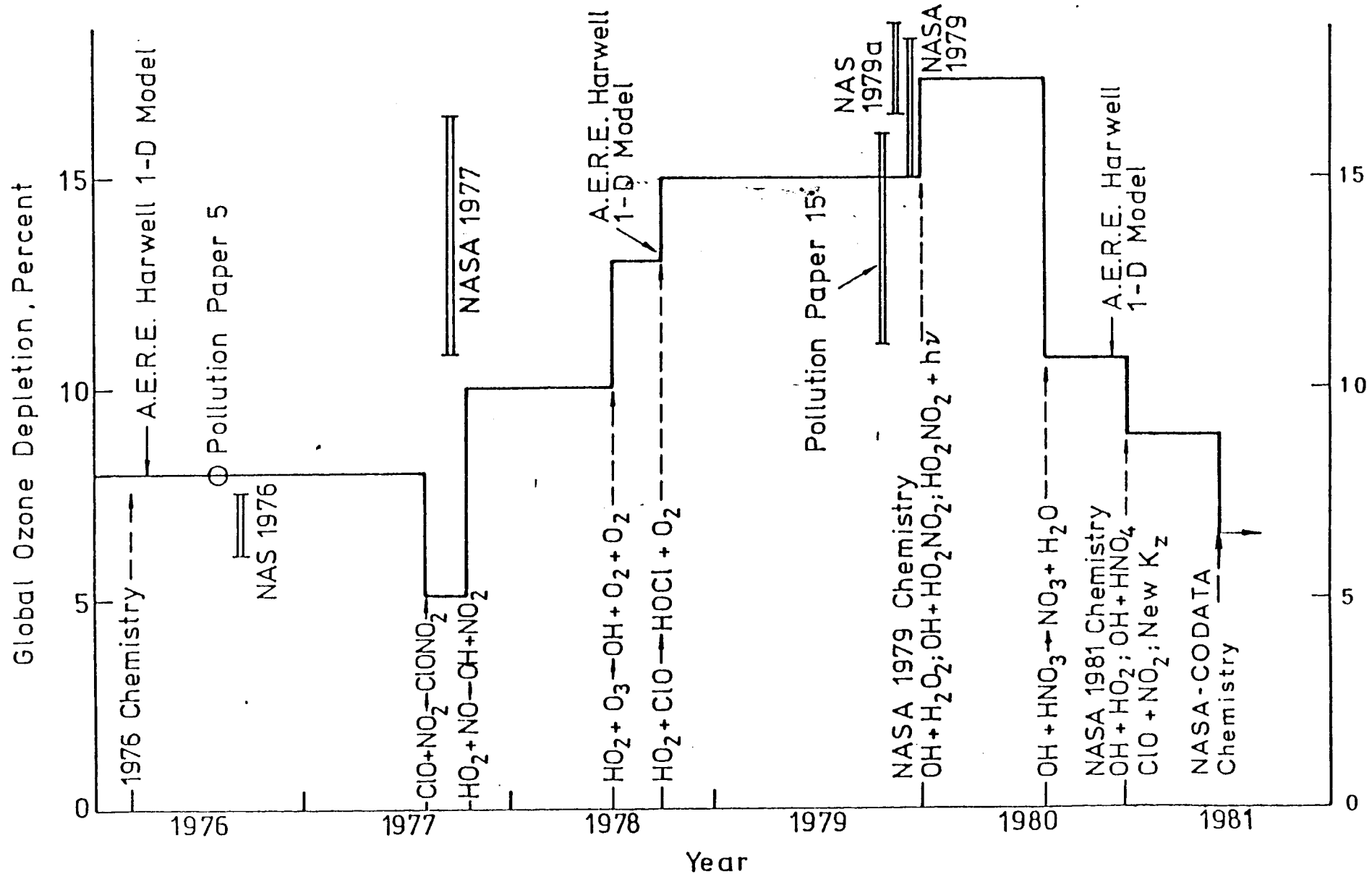


FIGURE 1 The possible steady state global ozone depletion due to the continued release of CFCs 11 and 12 at the 1976 production rates calculated with the AERE Harwell 1-D model. The figure shows the effects of the major re-evaluations in the chemical kinetic input data and the ozone depletion estimates given in the major reports on the stratospheric ozone layer.

error limits of such measurements. Thus available atmospheric data do not provide the means of validating atmospheric models with the accuracy required and considerable improvement is needed both in coverage and precision.

From an examination of the behaviour of halocarbons and reactive chlorine in a one-dimensional photochemical model of the stratosphere it has proved possible to construct simplified models which faithfully reproduce the predicted behaviour in the more complex models. In this way, it is possible to evaluate the effect of complicated patterns of halocarbon usage on the stratospheric ozone amount from the results of a few experiments with the more complex one-dimensional model, with a large saving in computer time.

The HARWELL 2-D model has been used to investigate possible ozone production in the troposphere as a result of emissions of NO<sub>x</sub> from subsonic aircraft<sup>(12)</sup>. The effect is found to be more marked in the northern hemisphere than the southern hemisphere and most noticeable in the summer upper troposphere. The increase in total column ozone at 30°N (annual mean) is calculated to be ~ 0.3% on the basis of 1975 aircraft NO<sub>x</sub> emissions ( $4.9 \times 10^8$  kg NO<sub>2</sub> yr<sup>-1</sup>) with a local increase of 2% between 8 and 10 km altitude at 30°N in summer. This calculated ozone increase may well have largely offset the present day predicted decrease in total column ozone due to CFCs 11 and 12 in the northern hemisphere but to have only a minor effect on that in the southern hemisphere where the effects of aircraft emissions are smaller.

#### 4. Assessments

A review of the effects of man-made pollutants on the stratospheric ozone layer has recently been published<sup>(13)</sup> which assesses some of the changes which have occurred since the publication of the UK Dept of Environment's Pollution Paper No.15 (chlorofluorocarbons and their effect on stratospheric ozone).

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