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Authors

Prather, Michael
Garcia, Maria M
Suzo, Robert
et al.

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Global Impact of the Antarctic Ozone Hole: Dynamical Dilution With a Three-Dimensional Chemical Transport Model

MICHAEL PRATHER

NASA Goddard Space Flight Center, Institute for Space Studies, New York

MARIA M. GARCIA

Department of Applied Physics and Nuclear Engineering, Columbia University, New York

ROBERT SUOZZO

ST Systems Corp., New York

DAVID RIND

NASA Goddard Space Flight Center, Institute for Space Studies, New York

A study of the Antarctic ozone hole has been made with a three-dimensional chemical transport model (CTM) using a linearized photochemistry for ozone. The tracer model uses the winds and convection from the Goddard Institute for Space Studies general circulation model ($8^\circ \times 10^\circ \times 23$ layers). The general circulation model (GCM) develops an Antarctic circumpolar vortex in early winter with strong westerlies that reverse in austral spring; the circulation compares favorably with the observed climatologies. A 4-year control run of the CTM with annually repeating winds produces ozone distributions that compare reasonably with the observed climatology. We examine different linearizations of the ozone chemistry and show that the calculated column ozone is sensitive to the chemical time constants in the lower stratosphere. In separate numerical experiments a hypothetical Antarctic ozone "hole" is induced on September 1 and on October 1; the CTM is integrated for 1 year with a linearized model that assumes standard photochemistry, not including the heterogeneous reactions and unusual chemistry associated with formation of the ozone hole. The initial depletion, assumed to be 90% of the O_3 poleward of $70^\circ S$ between 22 and 200 mbar, amounts to about 5% of the total O_3 in the southern hemisphere. As the vortex breaks down and the ozone hole is dispersed, significant depletions to column ozone, of order 10 Dobson units (3%) occur as far north as $40^\circ S$ during austral summer. One year later, only 30% of the original depletion remains, mostly below 100 mbar and poleward of $30^\circ S$. The October 1 initialization is continued for a second year, the ozone hole being reinduced 1 year later with the same parameterization. The cumulative effects from the year before are noticeable but add only 20% to the depletion. A budget analysis for the southern high-latitude stratosphere (10 - 350 mbar $\times$ 31° - $90^\circ S$) indicates the ozone hole is replenished equally by photochemical regeneration and by reduced transport of ozone into the troposphere, with a lesser fraction being filled in by an increased flux from the tropical stratosphere.

1. INTRODUCTION

Observations of ozone over Antarctica have shown a decrease of about 50% in the column abundance during austral spring since the late 1970s [Farman *et al.*, 1985; Stolarski *et al.*, 1986]. Concern over this depletion has led to several major expeditions in order to understand the chemistry of the Antarctic stratosphere: National Ozone Expedition [see Solomon *et al.*, 1987] and Airborne Antarctic Ozone Experiment [see Tuck *et al.*, 1988; Anderson *et al.*, 1988; Tuck *et al.*, 1989]. These field studies have implicated high levels of chlorine as being associated with, and possibly as being an immediate cause of the springtime ozone decline. Global satellite observations of the column abundance of ozone since 1979 [Total Ozone Mapping Spectrometer (TOMS): Krueger *et al.*, 1988] now clearly indicate a detectable decline in

column ozone throughout the year, even as far from the Antarctic continent as $50^\circ S$ (International Ozone Trends Panel Report, see Watson *et al.* [1988]). Many models have been proposed for the chlorine-induced losses within the Antarctic vortex in the early spring [Solomon *et al.*, 1986; McElroy *et al.*, 1986; Crutzen and Arnold, 1986; Molina and Molina, 1986; Cox and Hayman, 1988; Rodriguez *et al.*, 1988; Wofsy *et al.*, 1988], but these theories do not account for the observed hemisphere-wide, year-round depletion.

It has been widely suggested that once the Antarctic ozone hole is photochemically generated in the spring the ozone deficit remains in the stratosphere throughout the year, influencing extrapolar latitudes, and contributing to the formation of the ozone hole in the following years. This hypothesis is examined here using a three-dimensional (3D) chemical transport model (CTM) for the stratosphere with linearized ozone chemistry. Early work on ozone in a 3D CTM was done at the Geophysical Fluid Dynamics Laboratory (GFDL) [Mahlman *et al.*, 1980; Levy *et al.*, 1985]. More recently, the application of linearized chemistry in a general

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circulation model has been demonstrated by *Cariolle and Déqué* [1986].

On the basis of observations we impose an Antarctic ozone hole and then follow the dynamical dilution of the ozone-depleted air for the next 12 months. We assume that the unusual chemistry responsible for formation of the ozone hole has disappeared and no longer influences the subsequent evolution. Similarly, we assume that the general circulation is unchanged, and thus we do not include the predicted feedback mechanism by which the presence of the ozone hole helps to isolate further the Antarctic vortex [*Kiehl et al.*, 1988]. The initial ozone deficit is conserved by dynamical transports and can be eliminated only through photochemical regeneration in the stratosphere or mixing into the lower troposphere where ozone is effectively removed near the surface.

The 3D CTM calculations presented here show that transport of ozone-poor air from the Antarctic vortex results in measurable decreases to column ozone (>1%) extending to 30°S during austral summer. Approximately 70% of the initially prescribed ozone deficit is replenished through stratospheric chemistry by the end of the year. Year-to-year accumulation of residual deficits is shown to play a minor role at best in the rapid decline of Antarctic ozone each October, as observed over the past decade. These results are similar to 2D model simulations also presented at the Polar Ozone Workshop [*Ko et al.*, 1988].

The dynamical and chemical tracer models are described in section 2. The linearized chemistry is based on observed ozone, temperature and trace gas climatologies for the present atmosphere. Alternate treatments of the linearization and the resulting ozone distributions for standard climatologies are presented in section 3. The CTM is derived from the general circulation model (GCM) for the middle atmosphere developed at the Goddard Institute for Space Studies (GISS) [*Rind et al.*, 1988a, b]. The Antarctic meteorology (winds, temperatures) from the GCM is described in section 4 with an emphasis on the breakup of the winter vortex. Results from the simulated evolution of the ozone hole are given in section 5, and conclusions in section 6.

2. CHEMICAL TRANSPORT MODEL

The chemical transport model solves the continuity equations for a set of chemically reactive tracers over a 3D grid. This task involves computing the changes in tracer distribution due to advection by winds, convective and diffusive mixing (troposphere only), surface emissions and uptake, and in situ chemical production and loss. Details of the CTM are described by *Prather et al.* [1987], who documents the nine layer tropospheric version of the CTM. The major difference between the tropospheric version and the 21-layer stratospheric model used here is in the vertical structure described below. The stratospheric CTM has been used before in a study of meteoric sources [*Prather and Rodriguez*, 1988] and in the NASA 2D intercomparison [*Jackman et al.*, 1989].

The CTM uses the wind fields from the 23-layer stratospheric version of the GISS general circulation model [*Rind et al.*, 1988a, b]. A simulated 1-year climatology of the stratosphere and troposphere from the GCM are stored on history tapes, containing 8-hour averages of the wind and pressure fields plus monthly averages of temperatures and convective patterns. The CTM shares the grid resolution of

the stratospheric GCM shown in Table 1; it stores and recalculates the total mass of air and tracer within a volume or "grid box," designated in (longitude, latitude, pressure) coordinates by (I, J, L) and defined by the boundaries given in Table 1. The horizontal resolution of the CTM is 7.8° in latitude and 10° in longitude; vertical resolution within the stratosphere is approximately 5 km.

The CTM computes advective transport of a tracer using an upstream method that conserves the second-order moments of the tracer distribution subject to the constraint of positive definite tracer concentrations [second-order moments (SOM) with limits: *Prather*, 1986]. The SOM method conserves tracer to arbitrary (computer limited) accuracy, is highly accurate and nondiffusive but requires nine moments of the tracer distribution to be stored within each grid box. The additional expenditure in storage is more than compensated for by the increase in effective spatial resolution. The SOM method partially compensates for the coarse vertical resolution of the GCM, is able to maintain steep vertical gradients in tracer mixing ratio of more than a factor of 10 between adjacent layers but cannot make up for the poor resolution of the tropopause and lower stratosphere in this version of the GCM.

3. OZONE CHEMISTRY

Odd Oxygen and Chemical Steady State

The photochemical reactions that define the abundance of ozone in the stratosphere are often written in terms of the family odd oxygen,

$$O_x = O_3 + O(^1D) + O(^3P) \quad (1)$$

This definition of O_x is convenient for understanding the chemical terms that control ozone because several reactions lead to rapid exchange between members of the O_x family, but to no net change in odd oxygen.

(We use $h\nu$ to denote a photon and M to denote another molecule, usually N_2 or O_2 .) Ozone is the principal member of the O_x family at altitudes below 50 km, and thus the chemically induced rate of change of the ozone mixing ratio, f_{O_3} (vol/vol), may be approximated in terms of O_x ,

$$df_{O_3}/dt \approx df_{O_x}/dt = [P-L]_{O_x} \quad (5)$$

where $[P-L]$ denotes net production (vol/vol mixing ratio per second), including both production and loss terms.

Overall there are more than 30 reactions involving odd oxygen species that contribute directly to the net production [*DeMore et al.*, 1987]. The value of $[P-L]$ can be calculated rigorously under given stratospheric conditions, but the assignment of individual reactions to production (P) or loss (L) is often ambiguous. For example, the reaction

TABLE 1. CTM Coordinates for 23-Layer GCM

Index <i>I</i>	Longitude, deg	Index <i>J</i>	Latitude, deg	Index <i>L</i>	Pressure Level, mbar	Approximate Altitude, km
1	175E	1	90.0S	21	0.002	91
2	175W	2	86.1S	20	0.021	74
3	165W	3	78.3S	19	0.047	69
4	155W	4	70.4S	18	0.10	64
5	145W	5	62.6S	17	0.22	63
6	135W	6	54.8S	16	0.46	53
7	125W	7	47.0S	15	1.00	48
8	115W	8	39.1S	14	2.15	43
9	105W	9	31.3S	13	4.64	37
10	95W	10	23.5S	12	10.0	32
11	85W	11	15.7S	11	21.5	27
12	75W	12	7.8S	10	46.4	21
13	65W	13	0.0	9	100	16
14	55W	14	7.8N	8	203	11
15	45W	15	15.7N	7	346	7.4
16	35W	16	23.5N	6	507	4.7
17	25W	17	31.3N	5	664	2.8
18	15W	18	39.1N	4	797	1.6
19	5W	19	47.0N	3	884	0.9
20	5E	20	54.8N	2	929	0.5
	15E	21	62.6N	1	960	0.3
	•	22	70.4N		984	0.0
	•	23	78.3N			
34	145E	24	86.1N			
35	155E		90.0N			
36	165E					
	175E					

GCM layers 21-23 are combined into CTM layer 21.

leads to no net change in O_x if it is followed, as is often the case, by

where O represents either form of atomic oxygen.

The primary reaction responsible for generating the ozone layer, photolysis of molecular oxygen, is clearly assignable to production of odd oxygen.

Reactions involving peroxy radicals can lead to increases in O_x ,

but are more difficult to attribute directly to production of O_x unless the definition of O_x is extended to include NO_2 .

Loss of odd oxygen in the stratosphere occurs through many different reaction sequences, in particular, through catalytic cycles,

where X may be either NO or Cl. For these pathways, loss of odd oxygen will not respond in a simple linear manner to changes in ozone.

The chemical model used in these calculations is restricted to gas-phase, homogeneous reactions. Moreover, the residual impact of heterogeneous chemistry, notably the repartitioning of chlorine into ClO, has not been included. The linearization of the ozone chemistry for the CTM assumes that the abundances of the chemical families and their partitioning are unperturbed by polar stratospheric clouds. This is clearly not the case over Antarctica where extensive loss of NO_y and H₂O have been observed. Both NO_y and H₂O are long-lived in the lower stratosphere, and their depletion would be propagated and diluted into the mid-latitude stratosphere as shown here for ozone. Possible chemical impacts of denitrification on mid-latitude ozone are examined in a parallel paper [Prather and Jaffe, 1990] and discussed in the conclusions.

The observed concentration of O₃ in the stratosphere, f^o , is rarely in local chemical steady state (defined by $df^{ss}/dt = [P-L]^{ss} = 0$), except in the upper stratosphere where chemical time constants are short relative to dynamical time scales. In the tropical lower stratosphere, upward transport brings in tropospheric air with low mixing ratios of ozone. Under these conditions, ozone is being produced more rapidly than it is destroyed: $[P-L]^o > 0$, and $f^o < f^{ss}$. At mid and high latitudes, the reverse is true with predominantly downward transport of air from the upper stratosphere containing high mixing ratios of O₃. Loss exceeds production, and $f^o > f^{ss}$.

Linearization for the CTM

The CTM is not currently capable of solving directly for the net chemical tendency of ozone using the complete photochemical model (e.g., see discussion of Cariolle and Déqué [1986]). Such a calculation would require evaluation of the species concentrations and kinetic rates over diurnal cycles, as well as simultaneous transport of many trace gases along with ozone. For ozone in the current CTM we use a linearized chemical model for ozone derived from a 1D model with complete photochemistry [Logan et al., 1978; Prather et al., 1984; DeMore et al., 1987]. We use observed climatologies to define profiles for the trace gases and families: O₃, O₂, CH₄, H₂O, H₂, CO, NO_y (= HNO₃ + NO + NO₂ + NO₃ + N₂O₅ + HNO₂ + HO₂NO₂ + ClONO₂), Cl_x (= HCl + ClO + Cl + Cl₂ + HOCl + ClONO₂ + (ClO)₂), Br_x (= Br + BrO + HBr + HOBr + BrONO₂ + BrCl). The values for O₃ and temperature (and hence, O₂ and N₂) are taken from standard climatologies for the monthly, zonal means: 12 months by 10° in latitude by 2 km in altitude [see McPeters et al., 1984]. These quantities define the ultraviolet radiation field and hence the key photolysis rates. The profiles of the trace gases are based on a single average of mid-latitude measurements plus model results and are shifted in altitude according to the tropopause height appropriate for a given latitude and month (see Table 2, also Logan et al. [1978]).

The linear model for ozone photochemistry describes the tendency in ozone mixing ratio (vol/vol per second) for perturbations about the current state of the stratosphere (f^o). It does not take into account large, nonlinear perturbations beyond the climatological mean state; it does not include the heterogeneous chemistry associated with formation of the Antarctic ozone hole. Cariolle and Déqué [1986] present a linearization that includes in addition the effects of temperature

perturbations and of overhead ozone columns. Their chemical model is based on the distribution of ozone and other trace gases from their 2D model and is directly coupled with their GCM.

In contrast we choose to linearize the ozone chemistry solely in terms of the ozone mixing ratio by expanding $[P-L]_{O_3}$ to first order about the chemical conditions of the observed atmosphere.

$$[P-L] = [P-L]^o + (f-f^o) d[P-L]/df + \text{order}(f-f^o)^2 \quad (12)$$

It is convenient to express this formula in terms of a linear projection of the mixing ratio at $[P-L] = 0$, denoted by f^* , and a time constant, T .

$$f^* = f^o - [P-L]^o / (d[P-L]/df) \quad (13)$$

$$T = -1 / (d[P-L]/df) \quad (14)$$

The projected steady state mixing ratio is based on a linear extrapolation about the observed state and is therefore not likely to be the true steady state value. For any mixing ratio f , we calculate the net production of ozone from

$$[P-L] = (f^* - f) / T \quad (15)$$

Our derivation of f^* and T depends on how we choose to interpret the derivative $d[P-L]/df$.

We have examined three different forms for the linearization of the ozone chemistry, designated X, Y and Z. Chemistry X assumes that stratospheric ozone is everywhere in chemical steady state ($f^o = f^*$) and that photolysis of molecular oxygen (reaction (8)) is the only source of odd oxygen ($T = f^o/P^o$). These assumptions will clearly introduce errors in the lower stratosphere. Nevertheless, chemistry X has a certain simplicity in that the model depends only on the values of ozone and temperature (i.e., photolysis of O₂) and not on the assumed distribution of trace gases or other details of the chemistry.

Chemistry Y uses the full set of photochemical reactions to calculate the value of $[P-L]^o$ and assumes that primary production of odd oxygen, P^o , can be defined by reactions (8)-(10) above. We assume that this production term is invariant with respect to changes in the local concentration of O₃, and that the loss term, varying linearly for small perturbations about f^o , can be defined from the residual

$$L^o = P^o - [P-L]^o \quad (16)$$

The projected steady state mixing ratio and time constant for ozone under these conditions are related by

$$T = f^o / L^o = f^* / P^o \quad (17)$$

Chemistry Z is defined by the linearization described in equations (12)-(14). The derivative term is evaluated numerically in the photochemical model by varying the local concentration of ozone about the observed value. Figure 1 shows the time scale for ozone (T) during January using both chemistries X and Z. The values of T derived from chemistry X correspond to the ozone replenishment times used by Solomon et al. [1980] which are much slower than the actual chemical time scales for ozone.

TABLE 2. Assumed Profiles for Ozone Chemistry

Altitude, km			NO _y , ppb	Cl _x , ppb	Br _x , ppt	CH ₄ , ppm	H ₂ O, ppm	H ₂ , ppm	CO, ppb	C ₂ H ₆ , ppt
5°-25°	35°-55°	65°-85°								
62	58	54	11.0	2.60	20	0.13	5.94	0.50	15	0
60	56	52	12.0	2.60	20	0.13	5.94	0.50	15	0
58	54	50	13.0	2.60	20	0.14	5.92	0.50	15	0
56	52	48	13.0	2.60	20	0.15	5.90	0.50	15	0
54	50	46	14.0	2.60	20	0.16	5.88	0.50	15	0
52	48	44	15.0	2.60	20	0.17	5.86	0.50	15	0
50	46	42	15.0	2.55	20	0.19	5.82	0.50	15	0
48	44	40	16.0	2.55	20	0.22	5.76	0.50	15	0
46	42	38	17.0	2.55	20	0.25	5.70	0.50	15	0
44	40	36	18.0	2.50	20	0.30	5.60	0.50	15	0
42	38	34	18.0	2.50	20	0.35	5.50	0.50	15	0
40	36	32	19.0	2.50	20	0.42	5.36	0.50	15	0
38	34	30	20.0	2.45	20	0.50	5.20	0.50	15	0
36	32	28	19.0	2.40	20	0.60	5.00	0.50	15	0
34	30	26	18.0	2.30	20	0.70	4.80	0.50	15	0.01
32	28	24	17.0	2.20	20	0.80	4.60	0.50	15	0.06
30	26	22	16.0	2.04	20	0.95	4.30	0.50	15	0.40
28	24	20	14.0	1.80	20	1.05	4.10	0.50	15	2
26	22	18	12.0	1.50	20	1.15	3.90	0.50	15	10
24	20	16	8.0	1.16	20	1.25	3.70	0.50	15	40
22	18	14	6.0	0.80	20	1.40	3.40	0.50	15	160
20	16	12	4.0	0.45	20	1.50	3.20	0.50	15	480
18	14	10	2.0	0.15	20	1.60	3.00	0.50	15	1100
16	12	8	0.5	0.11	1	1.60	4.00	0.50	50	1200
14	10	6	0.3	0.10	1	1.60	144.	0.50	100	1300

We use the 1D photochemical model, as described above, in conjunction with the assumed climatology for ozone, temperature and trace gases to calculate tables of f^* and T that are averaged vertically over each stratospheric layer in the CTM. The steady state mixing ratios for ozone (either projected or actual using chemistry Y and Z) have a problem that is shared with most current photochemical models: predicted values are 10-30% below those observed over the altitude range 1-10 mbar. This discrepancy represents a fundamental, systematic difference between models and measurements that cannot be resolved here. These differences occur in the upper stratosphere where the time constant to achieve a steady state is rapid, and thus the observed values should represent a true photochemical steady state which is not perturbed by transport. We have chosen therefore to use the observed mixing ratios as the projected steady state values where the time constant, T , is less than 7 days and to interpolate smoothly between f^o and f^* for T greater than 7 days.

The net chemical change for ozone in each grid box of the CTM is based on the appropriate entry from these tables,

$$f_{O_3}(t) - f_{O_3}(0) = (f^* - f_{O_3}(0)) (1 - e^{-t/T}) \quad (18)$$

and is approximated for each 4-hour time step (T in seconds) as

$$f_{O_3}(t) = f_{O_3}(0) + (f^* - f_{O_3}(0)) \min[14400/T, 1] \quad (19)$$

In some instances, the projected steady state mixing ratios are negative. This result is not unphysical, because the projected values are not true steady state values but are only the linear extrapolation about the observed values. It occurs when the observed values are far from steady state and the time constant for O_3 is long, such as in the lower stratosphere at high latitudes. Negative values of O_3 are never reached because the time scale for chemistry is much longer than the time scale for circulation in the lower stratosphere.

The chemistry of tropospheric ozone differs substantially from that of stratospheric ozone and is not explicitly included in these calculations. Ozone is produced throughout the troposphere by the oxidation of NO (reactions (9) and (10)) and is destroyed predominantly in the lower troposphere by reactions involving water vapor, peroxy radicals, nonmethane hydrocarbons and surfaces. In these CTM simulations we ignore tropospheric production and mimic the photochemical and surface losses by setting the O_3 mixing ratio at the lower boundary to 20 ppb globally (layers $L = 1-3$).

Calculated Ozone Climatologies

A climatological simulation of the annual cycle of stratospheric ozone is computed for each of the chemical models. The same year of GCM winds and convective mixing (GISS reference no. 983N) is used repeatedly in the CTM to approach a steady state distribution for ozone (2 years for chemistries X and Y, 4 years for chemistry Z). The final year of these calculations provides both initial conditions and the

Fig. 1a

Fig. 1b

Fig. 1. Photochemical time scale (in days) for stratospheric ozone as a function of latitude and log-pressure for January using (a) chemistries X and (b) Z. The altitude coordinate in this and subsequent figures is $z^* = 16 \times \log_{10}(1000/p)$ where p is pressure (in millibar). Altitudes (0, 10, 20, 30, 40, 50, 60 km) correspond to pressures (1000, 237, 56, 13, 3.2, 0.7, 0.2 mbar), respectively.

control series for the simulations of the Antarctic ozone hole. These control runs are in approximate steady state, in so far as the mean annual cycle of ozone repeats to within about 1%.

In this section we examine the ozone climatology produced by the different chemical models (X, Y and Z). We compare with the Solar Backscatter Ultraviolet (SBUV) climatology used to develop the linearized chemistry for ozone. In the upper stratosphere our predicted O_3 should match the observed values, since chemical time constants are rapid and transport of ozone is unimportant. In the lower stratosphere, however, the ozone distribution is determined by a balance between chemical tendencies and transport, and the comparison provides a good test of our model. The Dobson map of observed column ozone is shown in Figure 2, and ozone mixing ratios from the SBUV climatology for January and April are given in Figure 3. The control run with chemistry-X produced concentrations of ozone in the lower stratosphere that were much smaller than observed in the tropics and yet much larger than observed at high latitudes. Calculated column ozone and profiles are given in Figures 4 and 5, respectively. The discrepancies between the chemistry X simulations and the observations indicate that the chemical tendencies for ozone in the lower stratosphere are too slow: both in producing ozone in the tropics and in destroying it at high latitudes. The problem lies in using the photolysis of molecular oxygen (reaction (8)) to determine the time constant for odd oxygen (see Figure 1). In the lower stratosphere this assumption underestimates significantly the production of O_x which is enhanced by reactions involving peroxy radical and NO

(reactions (9) and (10)). Furthermore, photolysis of O_2 is negligible at high latitudes in the lower stratosphere where chemical loss and transport are the predominant terms controlling ozone. Thus chemistry X predicts insufficient loss of ozone poleward of 40° in the lower stratosphere. The resulting Dobson map (Figure 4) shows this pattern clearly. (N.B. The early versions of the linearization, chemistry X and Y, used a coarse latitudinal grid for the chemical coefficients which may cause some registration errors in the contour intervals when comparing with the observed reference state, Figure 2.)

Chemistry Y corrects some of the first-order problems with chemistry X. Photochemical loss of ozone in the lower stratosphere is now accurately represented, and the time constants for odd oxygen are more rapid, giving greater chemical control over ozone throughout the lower stratosphere. Errors in the mixing ratios for the lower stratosphere are reduced; however, the calculated Dobson map still exaggerates the equator-to-pole gradient in column ozone (see Figures 6 and 7). Although the complete set of reactions for O_x are included, the chemical time scales are still too long (i.e., chemistry Y assumes that production and loss are defined linearly by several key rates, rather than correctly evaluating the coefficients by differentiating).

Our best model, chemistry Z, reproduces the observed column ozone to a globally averaged rms accuracy of 21% and overestimates global ozone by 10%. Use of the full chemical linearization in chemistry Z yields faster time scales for production and loss of ozone in the lower stratosphere. This

Fig. 2. A Dobson map of observed column ozone (Dobson units) as a function of season and latitude. The values are from the SBUV climatology, see text.

Fig. 3a

Fig. 3b

Fig. 3. Observed zonal mean ozone mixing ratios (ppmv) as a function of latitude and log-pressure (see Figure 1) for (a) January and (b) April. The values are from the SBUV climatology.

Fig. 4. A Dobson map of calculated column ozone using chemistry X.

version of the chemical model builds up the ozone column in the tropics and reduces it at high latitudes: a pattern closer to that observed. The calculated climatologies for ozone column and profiles are shown in Figures 8 and 9, respectively.

The calculated ozone column shows an increase from equator to pole along with a clear spring maximum and fall minimum at mid-latitudes, a pattern that is in basic agreement with observations. The tropical minimum moves from about 10°N in January to 10°S in August, similar to the observed seasonality. The tropical column amounts are about 20 Dobson units (DU) (8%) less than observed. The absolute values during the spring maximum are accurately reproduced, but the calculated fall minimum is not as deep, overestimating column ozone by 30%. Another obvious discrepancy in the model is the large gradient predicted in column ozone between 25 and 45 degrees latitude in both hemispheres. The observed gradients are equally large during spring and early summer but weaken by fall.

Calculated ozone mixing ratios agree with the observed climatology above 10 mbar where the chemical linearization forces the ozone concentrations to the observed climatology. In the tropics the major error in the calculated ozone lies between 10 and 50 mbar; the model underpredicts ozone concentrations by about 14%, accounting for most of the differences in the ozone column noted above. At mid and high latitudes the differences between model and observations show a seasonal pattern. The spring maximum, which is reproduced well in total column, shows small errors in ozone mixing ratios (i.e., less than or about 20%) with some cancellation between layers 10–11 (22–100 mbar) and layer 9 (100–200 mbar).

During the fall minimum the model significantly overpredicts the ozone mixing ratios between 100 and 300 mbar by about 0.5 ppmv (parts per million by volume). This large discrepancy can account for most of the error in predicting the fall minimum in column ozone and is likely caused by the CTM's circulation defining a stratosphere that extends too far into the troposphere for this season. In the southern hemisphere the spring maximum is overestimated by the model, due in part to the observations (being a time average of SBUV data for the period 1979–1987) including several years of the Antarctic ozone hole. This latter feature of the comparison with observations (1) is most apparent in the lower stratosphere, (2) may explain part of the discrepancies below 100 mbar even into the austral summer (see Figure 8, January), and (3) is consistent with the location of ozone deficits predicted after breakup of the hole (see section 5).

Globally, the annually averaged flux of ozone into the troposphere is 7.1×10^{10} molecules $\text{cm}^{-2} \text{s}^{-1}$. Seasonal variations in the stratosphere-to-troposphere flux are large, more than a factor of 3, with maxima in the northern hemisphere from October through March and in the southern hemisphere from May through September.

We wish to emphasize here that our ability to reproduce the observed distribution of ozone, given that distribution as input to the chemical model, is not trivial. Ozone in the middle and lower stratosphere is not in photochemical steady state. What we are testing here is a combination of CTM transport and photochemical tendencies for ozone in the lower and middle stratosphere. We show here that the photochemical tendency of ozone below 10 mbar is a critical element in determining

Fig. 5a

Fig. 5b

Fig. 5. Calculated zonal mean ozone mixing ratios (ppmv) as a function of latitude and log-pressure (see Figure 1) for (a) January and (b) April using chemistry X.

Fig. 6. A Dobson map of calculated column ozone using chemistry Y.

ozone concentrations in the lower stratosphere. Thus the GFDL simulations, using a chemical model to calculate ozone concentrations at 10 mbar and no chemistry below [Mahlman *et al.*, 1980; Levy *et al.*, 1985], would have overestimated ozone at mid-latitudes.

It should be noted that in these calculations the chemistry used in the CTM is not an *ab initio* calculation of ozone concentrations from first principles. As noted above, such models still have problems with the upper stratosphere. In summary, our errors in the simulation of ozone with chemistry Z could be due either to incorrect transport in the CTM, such as too rapid upwelling in the tropical lower stratosphere, or to errors in the chemical model. This question cannot be answered with the current experiments but will be examined in subsequent CTM simulations using other chemical tracers such as CH_4 , CFCl_3 , CF_2Cl_2 , N_2O and NO_y .

4. ANTARCTIC METEOROLOGY

How well does the model simulate the dynamic climatology of the Antarctic stratosphere, especially in the critical springtime months of September and October? A description of Antarctic meteorology for this time period is available from various studies using satellite observations [e.g., Harwood, 1975; Hartmann, 1976; Hartmann *et al.*, 1984; Mechoso *et al.*, 1985]. The transition from winter to summer tends to be zonally regular in the southern hemisphere; large longitudinal asymmetry in the zonal flow occurs less frequently (e.g., 1979 and 1988).

Between September and October the stratospheric jet moves downward and slightly poleward as zonal winds weaken in both the stratosphere and troposphere. The time-averaged zonal winds at subpolar latitudes switch from west to east in the upper stratosphere during the early part of November, and in the lower middle stratosphere during the latter part of November (see Figure 10a). During the years 1979-1984 the transition occurred between mid October and mid November in the upper stratosphere, and from mid November to as late as mid December in the lower stratosphere [Shiotani and Gille, 1987]. These basic features, as well as the changes in zonal wind for the rest of the year, are captured by the model's climatology, shown in Figure 10b. A more complete discussion of wind and temperature fields from the GCM is presented by Rind *et al.* [1988a, b].

Over Antarctica the transition from westward to eastward flow is indicative of the breakup of the polar cyclone in spring. This pattern is shown clearly in maps of potential vorticity from the GCM for the upper (1500 K surface, Figure 11a) and lower stratosphere (550 K surface, Figure 11b). In Figure 11 we have chosen dates ranging from October 6 to October 31 that span the breakup of the polar cyclone; results are specific to the year of winds used in the current CTM experiment, not from the 3-year climatology [Rind *et al.*, 1988a]. In the upper stratosphere the isolated polar vortex is associated with high values of potential vorticity present on October 6; it starts to dissipate by October 11; and the final spring warming occurs by October 16. In the lower

Fig. 7a

Fig. 7b

Fig. 7. Calculated zonal mean ozone mixing ratios (ppmv) as a function of latitude and log-pressure (see Figure 1) for (a) January and (b) April using chemistry Y.

Fig. 8. A Dobson map of calculated column ozone using chemistry Z.

stratosphere the pattern is similar but is delayed about 2 weeks. High values of potential vorticity present at the beginning of October start to decrease by October 26, and the final breakup of the vortex occurs by October 31.

The breakup of the Antarctic circumpolar vortex in October is driven both by solar heating and by eddy transport of heat and momentum. In addition to providing an accurate depiction of the mean fields and their changes, it is important that the model includes realistic eddy forcing. In Figure 12 we compare the divergence of the Eliassen-Palm flux in the model with that derived from observations for the southern hemisphere in September (typical of other winter months). The observations show deceleration of the westerlies (i.e., negative values) throughout much of the stratosphere and troposphere. An exception is the region of westerly acceleration from 60°S to 70°S in the stratosphere. The model reproduces the general appearance of these features although the maximum deceleration near the stratopause occurs in mid-latitudes rather than the subtropics. The magnitude of eddy forcing in the model is similar to that observed during late winter and early spring, considering the uncertainty in deriving the observed eddy fluxes.

This comparison of zonal fields of stratospheric winds and Eliassen-Palm flux indicates that the transport of chemical tracers averaged over time and longitude (i.e., the residual circulation, see *Andrews et al.* [1987] in the model may be similar to the transport occurring in the southern hemisphere during the breakup of the Antarctic ozone hole. The

agreement of these zonally averaged meteorological quantities does not necessarily imply that the transport of chemical tracers will be accurately simulated in three dimensions during this transient phenomenon when the winds reverse. Indeed the predictions of the dispersal of the ozone hole in this paper provide a test of the transport of trace species in the CTM. In conclusion, our average climatological model for the southern hemisphere may be representative of Antarctic spring, but further, detailed studies and/or simulations of the observations from individual years will be necessary to test the model.

5. BREAKUP OF THE ANTARCTIC OZONE HOLE

The focus of this paper is a 3D chemical tracer experiment in which we follow the dynamical dilution and chemical replenishment of the Antarctic ozone hole. The ozone hole in the model is formed by dividing ozone concentrations by a factor of 10 throughout the stratospheric region between 22 and 200 mbar (model layers $L = 9-11$) poleward of 70.5°S (model latitude bins $J = 1-3$). The spatial extent and the magnitude of ozone depletion are roughly consistent with observations [e.g., *Solomon and Schoeberl*, 1988] and were chosen for simplicity of initialization on the CTM grid. Following this arbitrary creation of the hole, the chemistry is assumed to be normal everywhere (i.e., chemistry Z; the simulations presented at the Polar Ozone Workshop in May 1988 used only chemistry X). The time of initialization is based on observations of the formation and breakup of the Antarctic ozone hole and is assumed to take place instantly on

Fig. 9. Calculated zonal mean ozone mixing ratios (ppmv) as a function of latitude and log-pressure (see Figure 1) for (a) January and (b) April using chemistry Z.

Fig. 10. Observed and modeled zonal mean winds (m s^{-1}) at 70°S as a function of season and log-pressure (see Figure 1). The observed winds (a) are taken from Wu *et al.* [1987], and model values (b) are based on the five-year climatology [Rind *et al.*, 1988a].

1500K POTENTIAL VORTICITY

Fig. 11a

550K POTENTIAL VORTICITY

Fig. 11b

Fig. 11. Potential vorticity ((a) $10^{-4} \text{ }^{\circ}\text{K m}^2 \text{ kg}^{-1} \text{ s}^{-1}$, and (b) $10^{-6} \text{ }^{\circ}\text{K m}^2 \text{ kg}^{-1} \text{ s}^{-1}$) from the GCM for the upper ($\Theta = 1500 \text{ K} \sim 40 \text{ km}$) and lower ($\Theta = 550 \text{ K} \sim 20 \text{ km}$) stratosphere of the southern hemisphere. Large negative values correspond to air within the polar vortex. Periods in October are chosen to illustrate the breakup of the circumpolar vortex. (Values are 5-day averages; date shown is end of averaging interval.) Model results are taken from the wind fields used in these CTM calculations.

October 1 before springtime breakup of the polar vortex in the CTM circulation (see section 4). The same year of GCM winds and convective mixing is used in both the control run and the ozone hole simulations. The ozone concentrations are integrated for two model years with the Antarctic ozone hole being reintroduced in the same manner for October 1 of the second year. We also performed a similar calculation for one model year, initializing the hole on September 1 as a test of the sensitivity of results to our initialization.

In describing the evolution of the Antarctic ozone hole we examine the deficit in ozone column relative to the control run (expressed in Dobson units or percent difference with respect to the control run). Figure 13a shows ozone column deficits (percent change) from the 2-year simulation beginning October 1; Figure 13b, from the 1-year run starting on September 1: One month after the experiment begins on October 1, deficits of order 8% are observed at 60°S , and reductions in column

ozone reach 1% as far north as 45°S . Ozone depletions spread rapidly through southern mid-latitudes by midsummer. By February 1 the deficit at 60°S declines to 6% while that at 30°S reaches 1%. Recovery of the ozone deficit continues almost uniformly through austral summer, slowing as winter approaches. By the following spring, column reductions at 60°S are only 2%, and more than two-thirds of the initial deficit has been regenerated by photochemical production of ozone. The calculation initialized on September 1 is almost identical to the October run, demonstrating the lack of sensitivity to these choices in initializing the ozone hole. Parallel model calculations (not shown) using chemistries X and Y produce similar results.

The cumulative buildup of an ozone hole through successive years of ozone depletion is found to be a second-order effect. Note that 1 month after reintroducing the hole for the second year, deficits at 60°S are of order 9.7% compared with 8% the

Fig. 12. Observed and modeled Eliassen-Palm flux divergencies (in meters per second per day) for September. Model results (a) are taken from the 5-year climatology [Rind *et al.*, 1988a] and (b) observations are taken from Mechoso *et al.*, [1985].

previous year. By January 1, deficits of order 1% could be found as far north as 25°S, as compared to 30°S in the first year. The average ozone depletion at the end of the second year is only 20% larger than that at the end of the first year, and for either year, amounts to approximately 0.7% of the global ozone content. Similar results, including both the latitudinal extent of mixing and the year-to-year accumulation of the Antarctic ozone hole, have been reported from calculations using more detailed chemistry in a 2D model [Sze *et al.*, 1989; Chipperfield and Pyle, 1988].

Changes in local ozone concentrations after the breakup of the Antarctic ozone hole are summarized in Figure 14, which shows the percent depletion of the zonally averaged mixing ratios relative to the control run for the months of January, April and July following the October 1 initialization. By the peak of austral summer the ozone deficit is displaced to lower altitudes and to mid-latitudes (Figure 14a, January). Ozone deficits of order 6% are found in the stratosphere between 50 and 200 mbar as far north as 40°S; deficits of order 4% reach 30°S; and part of the original depletion is seen in the troposphere. Passing through fall (Figure 14b, April) and into winter (Figure 14c, July) this trend continues: the magnitude of the ozone deficit decreases everywhere, and the region with greatest relative depletion continues to spread northward and downward into the troposphere. By winter, differences remaining in the altitude range 11–16 km (100–200 mbar) north of 40°S are only 4%. Most of the deficit that remains is distributed at lower altitudes below 350 mbar and south of 45°S. This seasonal trend is common to all simulations. In summary, stratospheric ozone depletion is transported into mid-latitudes and mixed into the troposphere as the ozone hole is dispersed, but there is a clear barrier to transport into the tropical stratosphere.

It is instructive to examine the processes controlling column ozone in the southern hemisphere, both with and without the Antarctic ozone hole. We choose to perform a budget analysis for the total amount of ozone in a "box" comprising the lower stratosphere (10 to 350 mbar, $L = 8$ –12) in the southern mid-latitudes (all longitudes poleward of 31°S, $I = 1$ –36 and $J = 1$ –8). The amount of ozone in this box on October 1 from the control run using chemistry Z is 83×10^{10} kg out of a global content of 332×10^{10} kg (349 DU). The cumulative change in this box through an annual cycle (Figure 15) is due to transport from the tropical stratosphere (denoted by 31°S), transport from the upper stratosphere (denoted by 10 mbar), transport into the troposphere (denoted by 350 mbar), and in situ chemical loss. The dominant source of ozone in the southern mid-latitude stratosphere is transport from the tropical stratosphere (62×10^{10} kg/yr); the loss is split equally between stratospheric chemistry and transport into the troposphere where atmospheric and surface reactions destroy ozone. The time constant to replace ozone in this stratospheric box is 1.2 years.

In the case of the ozone hole the budget analysis in Figure 16 plots the absolute difference with respect to the control run; the positive values represent replenishment of ozone. The imposed ozone hole is equal to an instantaneous loss on October 1 of 7.4×10^{10} kg, all within the southern mid-latitude stratosphere. Chemical loss is reduced in the presence of lower concentrations of ozone, and chemistry rapidly fills in more than 2×10^{10} kg during summer before becoming dormant by late fall for a total of 2.6×10^{10} kg. Similarly with less ozone, there is less transport into the troposphere, and this component (350 mbar) contributes uniformly throughout the following year a total of 2.6×10^{10} kg toward replenishing the ozone hole. There is a small increase over the year in the

Fig. 13a

Fig. 13b

Fig. 13. Percent differences in column ozone for the Antarctic ozone hole simulation relative to the control run in the southern hemisphere as a function of season and latitude using chemistry Z. The ozone hole was initialized on (a) October 1 of successive years for a 2-year calculation and (b) on September 1 for a 1-year calculation.

Fig. 14a

Fig. 14b

Fig. 14. Percent differences in zonal mean ozone mixing ratios for the Antarctic ozone hole simulation relative to the control run in the southern hemisphere as a function of log-pressure (see Figure 1) and latitude using chemistry Z. The ozone hole was initialized on October 1, and we show results for the months of (a) January, (b) April and (c) July. The dashed box represents the initial location of the ozone hole.

Fig. 14c

tropical source (0.7×10^{10} kg), but little change in the source from the upper stratosphere. By the end of the year the total increase in ozone within this box is 5.9×10^{10} kg, corresponding to replenishment of all but 20% of the initial deficit. An additional 10% of the original deficit (0.7×10^{10} kg) remains in the atmosphere but has been transported outside of the southern mid-latitude stratosphere. In particular, we predict significant decreases to upper tropospheric ozone in the southern hemisphere, of order 10% over most seasons, that are attributable to the Antarctic ozone hole.

6. CONCLUSIONS

In this study we have shown that the 3D chemical transport model produces an adequate, although not ideal, simulation of the observed ozone climatology using a simple, linearized model for chemistry. Inducing an Antarctic ozone hole and then returning the chemistry to its unperturbed state on October 1 gives a subsequent evolution of the ozone deficit that displays some of the observed features of ozone depletion in the southern mid-latitudes (TOMS observations; see also Atkinson *et al.* [1989]). This simulation predicts chemical regeneration of most of the ozone hole during the subsequent year with only modest residual accumulation after several years of recurring ozone holes.

In order to compare with observations during August and September, we must improve the initialization procedure, allowing for a more realistic generation of the ozone hole. The choice of initializing the ozone hole on October 1 ignores the photochemical destruction of ozone that is observed to

occur continuously since the return of sunlight in August. The initialization on September 1 is similarly unrealistic and only gives us some clues regarding the impact of ozone loss earlier in the season. Therefore the simulations shown here fail to account for ozone depletion within the vortex, or its dilution to mid-latitudes, that occurs in late winter and early austral spring.

Parallel work on the chemical propagation of the Antarctic ozone hole [Prather and Jaffe, 1990] has shown that additional ozone depletion is unlikely to occur during the transport of ozone-poor, chemically perturbed air parcels from the ozone hole to the mid-latitudes. Moreover, the transport of this denitrified air will reduce NO_y at mid-latitudes by 5-10%, which in turn leads to reduced chemical loss of ozone at mid-latitudes and a faster recovery from the effects of dilution of the ozone hole. On the other hand, the dilution and mixing of polar-processed air without ozone depletion may lead to additional loss at mid-latitudes. In general, the transport and mixing of chemically perturbed air parcels occurs on spatial scales much smaller than those resolved by the CTM but may be studied with the tracer model by transporting chemically perturbed air as a tracer with potential for ozone depletion.

Several problems with the ozone simulation point to directions of future research that would increase confidence in the CTM results. First, the control climatology of ozone must be used to study the role of vertical mixing, and particularly deep tropospheric convection, in controlling the fall minimum of column ozone at mid-latitudes. This discrepancy represents the most serious error in the current calculations. Second, the

Fig. 15. Ozone budget for the southern mid-latitude stratosphere from the control simulation using chemistry Z. The cumulative change throughout the year (10^{10} kg of O_3), beginning October 1, is shown for the box defined by all longitudes poleward of $31^\circ S$ between 10 mbar and 350 mbar (total ozone content = 83×10^{10} kg). The individual components include transport from the upper stratosphere (10 mbar, dotted curve), transport from the tropics ($31^\circ S$, long dash), transport into the troposphere (350 mbar, short dash), and in situ chemical loss (thick solid curve). The sum of these changes is also shown (thin solid curve).

Fig. 16. Differences in the ozone budget for the southern mid-latitude stratosphere between the control and the Antarctic hole simulation. See Figure 15 for definitions and legend. The cumulative changes are generally positive and represent the replenishment of ozone in this box by changes in the individual components of the budget from the simulation of the Antarctic ozone hole (initial loss = 7.4×10^{10} kg).

predicted interannual variability in ozone can be examined with the five different GCM model years currently stored on history tapes. Finally, we must continue validation of the CTM using other stratospheric tracers (N_2O and

chlorofluorocarbons studies are in progress) and model-model intercomparisons [e.g., Jackman *et al.*, 1989].

Development of the CTM will move toward a more realistic chemistry for ozone that, of necessity, includes simultaneous

transport of ozone, methane, water vapor and the families of odd nitrogen, chlorine and bromine. One immediate application of the linearized ozone chemistry developed here will be an interactive GCM/CTM simulation of the Antarctic ozone hole. In this calculation the transport and chemical replenishment of the ozone hole would be coupled with the radiative forcing and the dynamical response of the GCM.

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- R. Suozzo, ST Systems Corp., 2880 Broadway, New York, NY.
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-
- M. M. García, Columbia University, New York, NY.