Three-dimensional chemical model simulations of the ozone layer: 1979–2015

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SUMMARY

One-year simulations of stratospheric chemistry are performed in a general-circulation model (GCM). A fairly comprehensive description of stratospheric chemistry is included in a state-of-the-art GCM which has been extended to the middle mesosphere. The predicted ozone concentration is used in the model radiation scheme, thereby coupling the dynamical and chemical processes. Simulations commence on 1 March in each of the years 1979, 1994, 1999, 2004, 2009 and 2014, and consist of a 4 month spin-up period, followed by a 1-year integration. Past and projected values of halogen amounts and greenhouse gases (GHGs) are imposed on the model. The results for 1979–80 and 1994–95 are generally in good agreement with observations, indicating in the latter case a deep Antarctic ozone hole and some Arctic ozone loss. For the 1979 simulation only a very shallow ozone hole was simulated, in agreement with observations. In about the year 2005, the Antarctic ozone hole reaches its maximum size and globally averaged ozone reaches its minimum, depending on the month. Tropical ozone continues to decrease until about 2010. Results in the Arctic are dominated by interannual variability, but minimum ozone may not be attained until the year 2010. The results suggest that the increase in GHGs is delaying the onset of ozone recovery. Relative to 1980 conditions, the model changes in ozone result in small predicted increases in surface ultraviolet radiation in the Arctic and mid-latitude summer but large increases in the tropics and in the Antarctic summer.

KEYWORDS: Chemistry Climate Stratosphere Ultraviolet

1. INTRODUCTION

The ozone layer has a potential climate impact and provides protection at the surface from ultraviolet (UV) radiation, hence its depletion since the 1970s has been of great concern. As a result, international agreements have been forged to limit halogen emissions and it was concluded (e.g. WMO/UNEP (1995), chapter 6) that ozone depletion will continue to increase until the chlorine loading of the stratosphere reaches its maximum. Soon after, ozone is expected to start to recover. However, such predictions did not consider the effect of increasing greenhouse gases (GHGs). For example, the impact of a major radiative perturbation—doubling CO₂ amounts—had already been shown to enhance Arctic ozone depletion in some circumstances (Austin et al. 1992; Austin and Butchart 1994), because of reduced temperatures in the lower stratosphere which lead to increased heterogeneous ozone destruction on polar stratospheric clouds (PSCs). However, doubling of CO₂ amounts will not occur for a considerable time. Therefore, of more immediate interest is the likely behaviour of ozone over the next few decades and whether some interaction between chemistry and climate processes might occur on this shorter timescale.

Shindell et al. (1998) have recently addressed this, as well as the longer-timescale ozone behaviour, by integrating a low-resolution general-circulation model (GCM) with parametrized chemistry for the period 1959 to 2070. They showed that the effect of increasing GHGs was to increase the temperature gradient between the tropics and high latitudes in the stratosphere, which strengthened the zonal winds. In turn this led to reduced planetary wave amplitudes and a reduced frequency in the occurrence of sudden stratospheric warmings. Furthermore, these changes would occur even by the decade 2010–2020, so that the conditions identified by Austin and Butchart (1994) as leading to increased ozone depletion were satisfied. Consequently, the lowest ozone amounts would occur some 10 years after the peak in chlorine amounts. Part of the reason for this is that, although peak chlorine is expected to occur in about the year

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2000 in the stratosphere (see, for example, Montzka et al. (1996), SORG (1996) and WMO/UNEP (1999), chapter 11), high levels are expected to be maintained for many years before significant reduction occurs. Complicating the picture is the behaviour of bromine which, although subject to similarly stringent production quotas as chlorine, may be expected to be released more gradually into the atmosphere because of its use in fire extinguishers (see, for example, SORG (1996) and WMO/UNEP (1999), chapter 11 for discussions of the likely future emissions of brominated compounds).

The ECHAM GCM with more comprehensive chemistry has also been run for the future atmosphere by Dameris et al. (1998), but the model (Steil et al. 1998) has an upper boundary at 10 hPa which constrains the model meridional circulation (Austin et al. 1997; Rind et al. 1998). Results presented in WMO/UNEP (1999) chapter 12 for the ECHAM model and also for the ARPROBUS model (Lefèvre et al. 1994) show relatively small differences between the 1990s and 2015. Thus, there is currently considerable uncertainty concerning the timing and extent of future ozone recovery.

In this paper the behaviour of ozone during the recent past, and over the next few decades, is investigated further using the UK Meteorological Office (UKMO) Unified Model (UM) coupled with a stratospheric chemistry scheme. The model is integrated for 16 months starting 1 March in each of the years 1979, 1994, 1999, 2004, 2009 and 2014, with the first 4 months used as spin-up period to allow the model to come to an approximate steady state in the stratosphere. This provides annual snapshots of ozone behaviour without the full computational expense of a continuous multi-decadal simulation. The results are analysed to determine the likely minimum ozone amounts and the corresponding onset of ozone recovery, together with the impact on surface UV.

2. Model description

(a) General description

The UM is a GCM which was used in this study in extended form with an upper boundary in the middle mesosphere (0.1 hPa). The dynamical formulation and physical parametrisations were identical to those described by Butchart et al. (2000) and included sea surface boundary conditions from a coupled ocean–atmosphere simulation. In the version used here, the UM has a latitude–longitude resolution of 2.5° by 3.75° and there are 49 vertical levels, as shown in Fig. 1 of Austin et al. (1997). The model advected 15 tracers, and incorporated a stratospheric chemistry subroutine which was coupled to the model radiation scheme via the predicted ozone.

(b) Chemistry

A full range of chemical species and reactions, using rates from DeMore et al. (1994), were included to describe the key stratospheric families (O₅, HOₓ, NOₓ, CIₒₓ and BrO₂). The model also treated heterogeneous reactions on the surfaces of type I and type II PSCs, which were determined from thermodynamic considerations. Type I PSCs were treated as crystalline nitric acid trihydrate (NAT) (Hanson and Mauersberger 1988), and type II as water ice but, for simplicity, the PSCs were not sedimented in the model. In these simulations surface reactions on aerosols were not included. Short (60-day) integrations of the UM in this general configuration were described by Austin et al. (1997). However, in a preliminary longer integration for the 1994–95 case, model temperature biases, particularly in the middle stratosphere, were found to lead to excessive ozone depletion and it was found to be necessary to increase the temperatures by 5 K inside the heterogeneous chemistry subroutines. Nonetheless, it is currently common practice to adjust the temperatures in chemistry-climate models (for
example, IPCC (1999) chapter 4 ‘NASA* Goddard model’ and Steil et al. (1998)). This is discussed further in section 8.

To determine the concentrations of the long-lived species \( \text{H}_2\text{O}, \text{total nitrogen}, \text{total chlorine and total bromine}, \) the compact relationships between tracers noted by Plumb and Ko (1992) were exploited. In our previous work (Austin et al. 1997) this was done by advecting two tracers in the model, one of which (Tracer-1) was related to potential vorticity (initially equivalent latitude—Butchart and Remsberg (1986)) and the other (Tracer-2) was a vertically stratified tracer (initially potential temperature). For the longer timescale required here, the tracer advection scheme was found to be too diffusive, resulting in a considerable reduction in the horizontal gradient in Tracer-1 after 6 months. Therefore, the tracer was corrected every day during the integration (see the appendix). Also, because of model temperature biases the use of pressure rather than Tracer-2 was found to produce more realistic results, particularly for the Antarctic ozone hole. Hence, in the revised model used here, the long-lived tracers were parametrized in terms of Tracer-1 and pressure. Since the long-lived tracers control the shorter-lived species such as ozone, it is anticipated that any remaining errors in the advection scheme have no significant impact on the shorter-lived species in the model.

The chemical model operated between the tropopause and the lower mesosphere. Elsewhere, all the chemical constituents were advected passively, except for tropospheric ozone which was relaxed towards the initial conditions. At the ground the constituents were kept fixed and equal to the initial conditions to ensure that there was no net source or sink in the troposphere that would otherwise have become significant, particularly for ozone.

3. Integrations performed

Six integrations of approximately 16 months, each starting 1 March, were performed with the fully coupled model for conditions appropriate to 1979–80, 1994–95, 1999–00, 2004–05, 2009–10, and 2014–15. These cover the period of ozone decrease and expected recovery. Also, the 1979–1980 integration was extended by an extra four months to simulate the 1980 Antarctic spring period.

Meteorological initial conditions were taken from a sixty-year integration of a version of the model without chemistry (Butchart et al. 2000). For that integration and the integrations described here, the concentrations of the well-mixed GHGs \( \text{CO}_2, \text{CH}_4, \text{N}_2\text{O}, \text{CFC11} \) and CFC12 were specified according to the Intergovernmental Panel on Climate Change scenario IS92A (see Houghton et al. (1996) Table 2.5, and also Butchart et al. (2000) Fig. 1) and sea surface temperatures (SSTs) and sea ice amounts were taken from a Hadley Centre coupled atmosphere–ocean experiment ‘HadCM2’ (Mitchell et al. 1995; Mitchell and Johns 1997; Johns et al. 1997). Since the climate simulation of Butchart et al. (2000) was valid from only 1991 onwards, initial conditions for the 1979–80 experiment needed to be obtained elsewhere. The use of meteorological observations at the correct time may have led to major model drifts in the fields simulated by the coupled chemistry–climate experiment because of model bias. Therefore, for simplicity, the same initial conditions as for the 1994–95 integrations were used instead. The use of a four-month spin-up period reduced the effect that this may have had, and in subsequent analysis of the results the impact of the initial conditions was not noticeable.

The chemical initial conditions were taken from two-dimensional model results and shaped according to the local distribution of equivalent latitude, as described by Austin et al. (1997). Ozone data for the initial conditions were rescaled according to

* National Aeronautics and Space Administration.
TABLE 1. LONG-LIVED TRACER AMOUNTS USED IN THE MODEL SIMULATIONS

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<tr>
<td>Chlorine (ppb)</td>
<td>2.01</td>
<td>3.10</td>
<td>3.25</td>
<td>3.19</td>
<td>3.01</td>
<td>2.91</td>
</tr>
<tr>
<td>Bromine (ppt)</td>
<td>9.8</td>
<td>15.1</td>
<td>17.4</td>
<td>18.9</td>
<td>18.1</td>
<td>17.4</td>
</tr>
<tr>
<td>Effective halogen (ppb)</td>
<td>2.51</td>
<td>3.85</td>
<td>4.12</td>
<td>4.14</td>
<td>3.91</td>
<td>3.78</td>
</tr>
<tr>
<td>NO$_y$ (ppb)</td>
<td>22.4</td>
<td>23.4</td>
<td>23.8</td>
<td>24.1</td>
<td>24.4</td>
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Values are given at the equator for the top model level, except for NO$_y$, which is given at the peak (3.16 hPa) (units are parts per billion (ppb) or parts per trillion (ppt)). Other altitudes and latitude values are scaled in proportion to the 1994–95 values. The effective halogen is given by Cl$_y + 50.0 \times$ Br$_y$.

The distribution of Total Ozone Mapping Spectrometer (TOMS) data (Stolarski 1993). For the duration of each experiment the set of long-lived tracers remained constant as a function of equivalent latitude and pressure. For the individual experiments, the concentration of total nitrogen was made proportional to the N$_2$O value used in the radiation scheme. Similarly, total chlorine and total bromine were adjusted according to Table 1. The water vapour used in the chemistry scheme was kept the same for all the experiments. Although an increase has been observed (e.g. Oltmans and Hofmann 1995), global data are currently lacking and more information would be required before commencing a sequence of computationally expensive experiments with a specified H$_2$O rate of increase.

Chlorine amounts evolve in a similar way to past and current projections (WMO/UNEP 1999, chapter 11) although the model values are about 10–15% lower. However, the values imply a slower reduction than in previous compilations (e.g. SORG 1996). The bromine values are about 10% higher than are currently projected (e.g. WMO/UNEP 1999) and continue at higher levels for longer. As noted by Wamsley et al. (1998), previous aircraft measurements of total bromine were found to be biased high. Unfortunately, we were not aware of these adjustments to the measurements until after our numerical experiments had begun and, therefore, the slightly excessive bromine is projected into the future. Nonetheless, in terms of its impact on ozone depletion, this is probably a small correction. Equivalent halogen loading (see Table 1 and its footnote) is defined as the equivalent amount of chlorine which gives rise to the same level of ozone depletion. While this is a partially suspect concept, in that the multiplying factor for bromine is itself chlorine dependent, these values are also included in Table 1 for guidance. Note in particular that, with these values of chlorine and bromine, the continuing increase in the latter to 2010 and the slow reduction in the former after 2000 results in an equivalent halogen peak near 2005. The equivalent halogen level for 1979–80 is 2.51 ppb, about the level at which the Antarctic ozone hole formed (SORG 1996).

4. COMPARISONS WITH OZONE OBSERVATIONS

The general characteristics of the modelled ozone are indicated in Fig. 1. The model results are all lower than the observations, but only by about 15 Dobson Units (DU), and indicate a rapid change in the first few months, reaching stable values by early July. Thereafter the values show a clear annual cycle, with a slow downwards drift. The results show the same basic characteristics as the TOMS data, with a peak in northern hemisphere spring and a minimum in northern hemisphere summer. Also, the minimum observed in late October 1979 and early December 1996 occurred several months later in the model. During 1979–80 the model ozone was much higher than for 1994–95 which, in turn, was higher than for the other integrations. Some recovery in ozone appears to
have occurred in the model by 2015, but the results suggest a continuing reduction in ozone over at least the next 10 years.

Figure 2 shows the model results for (a) 1979–80 and (b) 1994–95 in more detail. In 1979–80 in the tropics, the model agrees very well with observations and is only about 10 DU less than the TOMS data. In the northern hemisphere, the model captures in part the steep gradient in the subtropics, leading into the ozone maximum at the pole during late spring. Near the pole, the peak model value was 437 DU on 5 March, some 38 DU lower than in the TOMS data for 26 March 1980. Also, in April, TOMS north polar ozone peaked at 477 DU on 8 April, but the model maximum was only about 407 DU at the pole. In the southern hemisphere, the modelled subtropical gradients are less than in the northern hemisphere, in agreement with observations, and the mid-latitude ozone peak is also well simulated. The main discrepancy in the southern hemisphere, is that the model simulates lower Antarctic ozone than observed and that this low ozone lasts longer. However, this may be mostly due to interannual variability since the observations show minima of 242 and 216 DU for 1979 and 1980, compared with model values of 203 and 230, respectively.

In the absence of observations for 1994–95, the data from 25 July 1996 to 24 July 1997 from the TOMS instrument on the Earth Probe satellite are compared with the results from the 1994–95 experiment in Figs. 2(b) and (d). As in the 1979–80 results many of the basic characteristics are reproduced in the model. At the north pole, the model value peaked on 5 April (405 DU) and 10 May (377 DU). In contrast, during 1996–97 total ozone was very low during much of the spring (275 DU at the end of March) and peaked on 1 May at 435 DU but, during the northern spring period, ozone depletion was exceptionally high compared with other recent years (Lefèvre et al. 1998). In the southern hemisphere, the model total ozone is generally in good agreement with the observations, particularly in the mid-latitude ozone maximum. The timing of the
Antarctic ozone hole is very well reproduced by the model but was not as deep as that observed in 1996.

5. Analysis of results: Temperature

(a) Global means

Analysis of the globally averaged temperatures (Fig. 3) further demonstrates that the model reaches approximate equilibrium after about 4 months of spin-up (cf. Fig. 1). As the greenhouse-gas concentrations increased, the model middle stratosphere became systematically colder. After 35 years, the 10 hPa temperatures had decreased by about 2 K. A similar cooling occurred in the lower stratosphere, but in the period December to June the lowest temperatures occurred in 2009–10, rather than 2014–15. The systematic change in globally and annually averaged temperatures is more clearly indicated in Fig. 4, where results from the transient climate-change experiment (Butchart et al. 2000) are also included out to the year 2030. In all cases, the temperatures diverge only slightly from a linear change and regression lines are drawn through the results. The temperatures from the chemistry simulations (grey lines) are about 1.5 K lower at 10 hPa and, during the period 1992–2015, are between 2.8 and 3.3 K lower at 46 hPa than for the transient-climate experiments (black lines). This is consistent with the chemistry model’s slight underprediction of ozone in comparison with current climatology used in the transient-climate experiment. Also, the rates of cooling at 10 hPa are almost identical.
Figure 3. Global mean temperature during the six calculations for the 10 hPa (upper panel) and 46 hPa (lower panel) levels. The months of the integrations, which were initialized on 1 March of the first year, are shown on the abscissa.

Figure 4. Global annual-mean temperature in the chemistry calculations (grey) compared with dynamics-only calculations (black) at (a) 10 hPa and (b) 46 hPa.

in the two models at about 0.53 K per decade. In contrast, at 46 hPa, the chemical model indicates approximately twice the cooling rate that occurred in the transient-climate experiment, implying that ozone changes have had a significant effect at this level (see also Ramaswamy et al. 1996). In particular, as ozone begins to show a recovery from about 2015 it might be anticipated that subsequent model temperatures will decrease at a much lower rate, or even show a continued increase as appears to have occurred between 2010 and 2015.
Figure 5. Ten-day running means of (a) the minimum temperature north of 45°N at the 46 hPa level for the six simulated northern winters, 1979, 1994, 1999, 2004, 2009 and 2014 (the shaded regions indicate the observed range in the UKMO data-assimilation analyses for 1991–92 to 1997–98 (Swinbank and O’Neill 1994)), and (b) the minimum temperature south of 45°S at the 46 hPa level for the same simulated southern winters (the shaded regions indicate the observed range in the UKMO data-assimilation analyses for 1992 to 1997).

(b) Wintertime

An important diagnostic to determine the likelihood of Arctic ozone depletion is the minimum temperature in the lower stratosphere north of 45°N indicated in Fig. 5 for the six experiments. For the earlier period (Fig. 5(a) upper panel) the results remain almost entirely within the 1990’s envelope of observations from the UKMO data assimilation analyses (Swinbank and O’Neill (1994), shaded region in Fig. 5). For the later period (Fig. 5(a) lower panel), the temperatures were again approximately within the 1990’s envelope, and during the early winter the minimum temperatures lower by about 1–2 K. In the spring, lowest temperatures were simulated in 1994 and again in 2009 with high temperatures in between (1999 and 2004). Hence, the results show considerable interannual variability for the Arctic, indicating the difficulty in detecting secular trends (see Butchart et al. 2000).

The equivalent comparison for the southern hemisphere winter is shown in Fig. 5(b). The model reproduced well the cooling in the early part of the winter (April–June), with all the calculations having minimum temperatures within the range of analysed values for most of the period. Minimum temperatures in the later period (Fig. 5(b) lower panel) were systematically lower than during the earlier period (Fig. 5(b) upper panel) during this time. In late winter and spring (July–October), temperatures were lower in the model than in the analyses, and showed no trend between the calculations. In the spring, a large part of this temperature bias resulted from a tendency for the model to delay the final warming; in all the calculations except 2014–15 no major warmings occurred before the
Figure 6. Zonally averaged total ozone (DU) simulated by the model minus the results from the experiment for 1994–95, for the years (a) 1999–00, (b) 2004–05, (c) 2009–10 and (d) 2014–15.

end of October. This shows that the winter vortex in the model is too cold and stable, as is found in many other GCMs.

6. ANALYSIS OF RESULTS: OZONE

(a) Ozone general features

Figure 6 shows the differences between the zonally averaged total ozone for the other four experiments, relative to the results for 1994–95. For the year 1999–00, throughout much of the northern hemisphere the model has lower ozone values than in 1994–95, but for a few days in the Arctic in March total ozone exceeded that for 1994–95 by over 40 DU. In the Antarctic some ozone depletion occurred during the Autumn in accordance with observations at this time of the year (Roscoe et al. 1997). The modelled Antarctic ozone hole was deeper than in 1994–95, and persisted longer, resulting in reductions of order 40 DU in November. In 2004–05 tropical ozone (Fig. 6(b)) decreased, while the results in northern high latitudes for April were similar to the year 2000. Again, the only region in the northern hemisphere where substantial increases in ozone occurred was in high latitudes in February and March. As in the results for 1994 and 1999, the ozone lasted into December. By the year 2010 there were generally lower ozone values than in the previous years in the northern hemisphere.
(Fig. 6(c)). In the southern hemisphere, significant recovery was evident, primarily in the ozone hole, but also in the band of high values in mid-latitudes. However, from July to January modelled Antarctic ozone was still lower in 2009 than in 1994. Finally, by the year 2014–15 the northern hemisphere started to show ozone recovery (Fig. 6(d)). Tropical values once more exceeded 225 DU everywhere while the region of positive ozone change in February and March in the northern hemisphere expanded. In the Antarctic the ozone hole recovered substantially, although small losses remained at other times of year.

Similar behaviour occurred in the model results for local minima (Table 2). In the north the minimum ozone was reached in 2010 and while ozone was not significantly lower in March than in 1995, the results for April indicate a much larger difference. In the south the minimum occurred in 1999 or 2004 depending on the month. Thus, the start of ozone recovery has become clear between 5 and 10 years earlier in the south than in the north, although with simulations every 5 years we cannot allow for interannual variability.

(b) Antarctic ozone hole

Since the chemical-depletion rate in the Antarctic ozone hole is a strongly nonlinear function of the halogen amount, it might be expected that significant recovery of spring Antarctic ozone may occur with a small decrease in halogen amounts. Further, the interannual variability in transport has a smaller impact. Thus, as suggested by the above results, the Antarctic ozone hole may offer the best prospect for an early detection of recovery of stratospheric ozone as halogen loadings fall.

Figure 7 shows the modelled October mean total ozone in the southern hemisphere for the six integrations between 1979 and 2014. For 1979, the model predicted a monthly-mean ozone minimum over the pole of just below 220 DU. The results for the 1980 ozone hole season (not shown), show higher ozone values (see, for example, Fig. 9), with the ozone minimum mainly associated with weak transport in the polar vortex rather than chemical depletion. A sharp contrast with the 1979 case can be seen in the model results for 1999 in which a deep ozone hole has developed, with minimum October mean total ozone values below 140 DU at approximately the time when stratospheric chlorine levels are expected to peak. Both 1994 and 1999 show markedly lower ozone in the mid-latitude ozone maxima that surround the ozone hole than in 1979. The results for 2004 show a similar level of ozone depletion as in 1999, although the mean ozone hole is larger and more elongated than before. Some degree of recovery appears to have occurred by 2009, with minimum values some 40 DU higher than in 2004, and the hole has remained elongated in shape. In 2014, recovery to within about 25 DU of the 1979 results, is indicated with significant increases in both the minimum values and the mid-latitude peak.
Figure 7. October mean total ozone (DU) for the integrations for the six years (a) 1979, (b) 1994, (c) 1999, (d) 2004, (e) 2009, and (f) 2014.
The local minimum column south of 60°S throughout the winter–spring period is shown in Fig. 8 (see also Table 2). The calculations for 1999 and 2004, around the time chlorine levels peak, both have seasonal minima of about 100 DU, whilst those for 1979 and 2014 both have minima above 170 DU. The 1994 and 2009 cases are intermediate between these groupings, with a minimum of about 130 DU. All the calculations showed the onset of ozone loss in September, when sunlight returns to southern high latitudes, and all except that for 2004 recovered by mid-December. In this case the polar vortex in the GCM persisted somewhat longer than for the other years. A measure of the size of the ozone hole is the area within the 220 DU contour, which is also plotted in Fig. 8. Again, the results in the years 1994–2009 show markedly distinct behaviour from the results in 1979 and 2014, with much larger areas. Also, despite an increase in minimum column ozone, the modelled ozone hole became larger in the decade 2000–2010 before decreasing in size. This effect may have been a result of cooling due to greenhouse-gas increases which would have increased the area where PSCs could form. In contrast, the minimum column ozone is related to the vertical range over which PSCs, and hence strong ozone depletion, can occur. Thus, small overall temperature decreases have had more of an effect at the lateral margins than at the top or bottom.

Figure 9 compares model predictions of minimum total ozone and maximum area within the 220 DU contour with observations from the TOMS series of instruments.
As before, model ozone levels in 1979 were about 20 DU below those observed, although values for 1980 were about 15 DU higher than those measured. However, the 1994 simulation was about 30 DU higher than TOMS, probably due to insufficient chlorine in the model (see Table 1). In general, the model has performed well both with and without ozone hole conditions. The model calculations for future states showed continual recovery in the minimum ozone, although this was not significantly above the current interannual variability until 2010. The overprediction of ozone loss in 1979 led to a small area within the 220 DU contour that was not observed. However, no values below 220 DU were obtained in the simulation of the 1980 southern spring. For 1994 and 1999 the model results were about 20% lower than current values. This may be due to the omission of aerosol heterogeneous processing above the NAT formation temperature. Nonetheless, the model results suggest that a further increase in the area of the ozone hole by some 15% will occur between 2000 and 2005.

The model simulations for 1979–2014 show that the estimation of the timing of the peak intensity and subsequent recovery of the Antarctic ozone hole depends on the quantity used to gauge its severity. In addition, the meteorological variability in the model calculations introduces uncertainty into such predictions. While the ozone loss rate during September between 12 and 20 km is a sensitive indicator of ozone depletion in the observations (Hofmann 1996), the impact of model vertical resolution and inaccuracies in the simulated vertical profile make this a less reliable diagnostic to analyse from the model. Nonetheless, the diagnostics presented here, such as the absolute minimum in total ozone and the maximum area in the 220 DU contour indicate that the ozone hole is likely to peak in the first decade of the next century, with recovery underway by 2010.

(c) Arctic ozone

The Arctic winter–spring is warmer and more variable than the Antarctic due to the greater influence of planetary waves. As a result, the frequency and duration of low temperatures at which PSCs form largely depend on the occurrence and timing of stratospheric warmings, as well as the final warming. This leads to a high degree of interannual variability in the amount of chemical ozone depletion, which makes an assessment of future trends in the Arctic much more uncertain than in the Antarctic.

In Fig. 10, the minimum total column ozone north of 60°N is shown for the northern winter–spring periods in the model integrations. All the calculations show
short-timescale variations of up to 50 DU due to transport. In addition, minimum ozone values increased from typically below 250 DU to about 300 DU during the period, depending on the timing of warmings. In the 2004–05 calculation for example, a strong warming occurred at the beginning of February, leading to increased transport and, therefore, a sharp rise in ozone values. In contrast, the 1994–95 calculation had no warmings before the final warming in April, and so ozone values increased more slowly. None of the results showed substantial decreases in minimum ozone column in February and March, however, as has been recently observed (see, for example, Lefèvre et al. (1998)). It is possible that some chemical ozone loss did occur in the calculations, but it is smaller than the ozone transport. Although the interannual variability is large in this region, the results indicate the first signs of ozone recovery by about 2015.

(d) Mid-latitude ozone

Annual mean mid-latitude total ozone amounts are shown in Fig. 11. In the north, simulated ozone was 20–30 DU less in total and the decrease, between 1980 and 1995, was about half that observed by TOMS. Future modelled ozone levels remained essentially unchanged from their value in 1995 at about 10 DU lower than in 1980. In the southern hemisphere, the model is in reasonable agreement with observations, with the 10 DU underprediction in the 1979–80 run within the range of interannual variability. The lowest ozone is predicted to occur in 1999–00 with slight increases thereafter. In this hemisphere, the change is probably dominated by transport from the ozone hole, a process well represented in the model. In contrast, in the northern hemisphere ozone depletion on aerosols, excluded in the current model, may be playing a more dominant role (Solomon et al. 1996; Jackman et al. 1996).

The largest reductions in column ozone occurred in the pressure ranges 200–70 hPa and 70–20 hPa. Although there was a general reduction in ozone amounts in the lower stratosphere, dynamical fluctuations masked this trend. In the northern hemisphere the ozone reductions between 1979 and 1999 were followed by smaller trends. In the southern hemisphere the ozone hole significantly perturbed the results. Smaller, but still significant, changes occurred above 20 hPa (about 27 km). Here, the lifetime of ozone falls to a few weeks or less, reducing the potential influence of ozone transport. The ozone evolved quite similarly in each hemisphere, and in the tropics (see section 6(e)), with a decreasing downward trend at 20–2 hPa, and a minimum in ozone above 2 hPa in
about the year 2000. At these altitudes the ClO + O catalytic cycle becomes a key ozone-loss process. Thus, as chlorine abundances rose between 1979 and 1994, ozone losses increased. Above 2 hPa (about 43 km), ozone is also lost by the O$_3$ + O reaction, which is particularly temperature sensitive. According to DeMore et al. (1994), the rate of this reaction at temperature $T$ is $8 \times 10^{-12} \exp(-2060/T)$ cm$^3$s$^{-1}$, implying a reaction rate change of $+3.3\%$ K$^{-1}$ at a typical temperature of 250 K. As this part of the atmosphere is predicted by the model to cool by about 1.0 K per decade, this corresponds to a 5% reduction in ozone loss by this reaction for the period 1999 and 2014. The modelled increase in ozone between these times is slightly smaller, at about 3%, since other loss processes with less temperature sensitivity (e.g. NO$_2$ + O) also operate.

(e) Tropical ozone

Figure 12 shows the modelled annual-mean tropical ozone, here used to denote the mean in the range 30°N to 30°S, which is about 15 DU lower than observed by TOMS. A more detailed statistical analysis of the data than can be presented here shows that the TOMS ozone has decreased in the latitude range 30°N–30°S by $-0.7 \pm 0.4 \%$ per decade (2σ, see Tables 4–6 in WMO/UNEP (1999) chapter 4.) for the slightly longer period to December 1997. This is equivalent to $3.0 \pm 1.6$ DU for the 15 years 1980–1995. Surface stations (see Tables 4–5 in WMO/UNEP (1999) chapter 4) give a larger decrease of $-1.1 \pm 0.3 \%$ per decade, equivalent to 4.5 ± 1.2 DU. The model results decreased by about 6 DU during the period 1980–1995, slightly larger than observed. In the model, tropical ozone continued to decrease until about 2010 before recovering slightly. Much of this change occurred in the pressure range 70–20 hPa, with a small change also occurring in the range 200–70 hPa. Below 200 hPa, ozone changes were very small, about 0.5 DU, since tropospheric ozone was relaxed towards constant values. The active chlorine, ClO$_x$ (= Cl + ClO + 2 × Cl$_2$O$_2$) present on a typical day (31 December) is shown in Fig. 13. Since these are diurnally averaged and include a period of darkness, peak daytime values are approximately three times the values indicated in the figure. In the upper stratosphere, the peak in ClO$_x$ occurred in 1994 but at lower levels the peak occurred later—between 10 and almost 20 hPa in 1999; at 20 hPa and below in 2009. Above 70 hPa the active chlorine is consistent with that which would be expected from gas-phase chemistry alone, while below 70 hPa, two peaks occurred due to heterogeneous processes occurring in the model. Investigation of the vertical variation of ClO$_x$ shows that the values are strongly anticorrelated with ozone in the pressure range 70–20 hPa, where the largest ozone column change occurred. Although the difference in the absolute magnitude of ClO$_x$ since 1980 is not high, this corresponds to a change in Cl mixing ratio, averaged over day and night, of about $1.5 \times 10^{-15}$, assuming a typical Cl–ClO mixing ratio from Brasseur and Solomon (1986). Given the reaction rate of Cl + O$_3$ of $8 \times 10^{-12}$ cm$^3$s$^{-1}$ (DeMore et al. 1994) and an ozone lifetime of about 50 days at 50 hPa (Brasseur and Solomon 1987), the approximate chemical ozone change is about 11% between 1980 and 2010. This is similar to the model computed value of 8%, suggesting that much of the change is due to increases in chlorine chemistry. Transport may also be playing a role but, unfortunately, with the current configuration of the model these processes cannot be separated. In the range 200 to 70 hPa there was further ozone loss, with heterogeneous chemistry playing a role, although this was significantly smaller than above 70 hPa. In this region of the atmosphere the model results are less reliable because of the possibility of spurious NAT formation near the cold tropical tropopause. Above 20 hPa model ozone changes were small and not clearly related to ClO$_x$ throughout the period. Of particular note,
however, are the results above 2 hPa which show an initial decrease due to the large increase in chlorine but from 1995 onwards, when chlorine changes were small, the ozone increased due to the decrease in upper stratospheric temperatures, as explained in more detail in section 6(d).

7. CLEAR-SKY SURFACE ULTRAVIOLET RADIATION LEVELS

Ozone provides an important protective shield against the harmful effects of UV radiation and here the impacts of the predicted ozone changes on the intensity of UV at the earth’s surface are presented. To simplify the analysis, clear-sky values are determined and we concentrate on erythema (sunburn) which has been implicated in increased incidences of skin cancer (e.g. Armstrong 1993).

UV irradiances were first computed for clear skies at noon using the radiation model used to compute photolysis rates. The irradiances were then weighted by the erythemal-action spectrum (McKinlay and Diffee 1987). When observed ozone is specified, the
Figure 14. The clear-sky erythemally weighted UV (mW m\(^{-2}\)) at noon in the simulations, as a function of month. (a) The monthly and regionally averaged UV for various latitude bands relative to the results for 1979–80, and (b) the maximum deviation from the daily zonal average attained during each month. Note the different scales on the ordinates of the graphs.
model was found to be in reasonable agreement with UV observations (Austin et al. 1999). Calculations are made for two conditions: firstly for mean UV levels in a latitude band and secondly for locally extreme values in that band.

Figure 14 shows the results obtained for high latitudes, mid latitudes and the tropics—see the figure caption for an explanation of the two quantities calculated. In the northern high latitudes the mean results show little difference between the simulations with a maximum increase (relative to the 1979–80 simulation) of 8 mW m\(^{-2}\) occurring in 1999–00. In summer, local values could be more than 30 mW m\(^{-2}\) higher than the regional average values of about 80 mW m\(^{-2}\). The southern hemisphere showed very significant features due to the change in behaviour of the Antarctic ozone hole. The regionally averaged values increased steadily up to 2005 and show, in particular, a predicted increase in early summer UV of some 35% relative to 1980, due to the extended duration of the ozone hole. Variability around the latitude circle was also very significantly affected and the results show that, during the early summer period, local UV can be more than double the zonal-average value. In contrast, from January to September, local UV extremes are calculated to be only about 25% of the peak summer values, similar to the values in the northern hemisphere.

Only small differences between the simulations occurred in mid-latitude ozone, as described previously, and this translates into small changes in mean UV of less than about 12 mW m\(^{-2}\) in the northern hemisphere summer relative to the 1979–80 simulation, compared with peak summer values of over 200 mW m\(^{-2}\). In southern middle latitudes the calculated UV changes were almost twice as large, and there were larger differences between the simulations. For most of the year the UV levels were calculated to be higher than in 1995. Extreme UV values for the northern mid-latitudes exceed the regional average by 90 mW m\(^{-2}\) in late spring but for southern mid-latitudes the influence of the Antarctic ozone hole is large, giving rise to local extreme values over 150 mW m\(^{-2}\) higher than the regional average.

Despite small differences in ozone, the model’s regionally averaged UV in the tropics showed a significant increase over the period 1980 to 2010, particularly in the northern-hemisphere spring when the increase was about 8%. This change is larger than any other region, except that occurring for a short period in high southern latitudes. Local extremes reached about 20% of the regional mean, with a clear annual variation peaking in northern-hemisphere summer. This annual variation tended to dominate the differences between individual years.

8. DISCUSSION

As indicated in section 6, the results show that the model provides good general agreement with observations for a wide range of conditions. Nonetheless, for a more accurate prediction of future ozone loss and recovery, there is a need to address several issues, as described below in subsections (a)–(c). In subsection (d) the results are compared with previous work.

(a) Temporal coverage of the results

The main limitation of this study stems from the use of one-year snapshots, with six snapshots covering a 35-year period, rather than using a continuous multi-decadal simulation. In the Antarctic the results are not significantly limited by the snapshot approach as the vortex is much less variable. However, in the Arctic, ozone changes in both the model and atmosphere are strongly dependent on the occurrence and timing
of stratospheric warmings. This also implies that detecting future ozone recovery in the northern hemisphere in the atmosphere will be a particular challenge.

(b) **Heterogeneous chemistry**

The results of the model simulations are strongly affected by the details of the parametrization of heterogeneous chemistry, but the composition of PSC particles remains uncertain. While a NAT-ice scheme is used here, certain observations seem to be in better agreement with a supercooled-ternary-solution model (Carslaw et al. 1994; Santee et al. 1998) of PSC formation. Other compositions (e.g. nitric acid dihydrate, sulphuric acid tetrahydrate, mixed phases) have also been proposed on the basis of laboratory and theoretical work (e.g. Middlebrook et al. 1992; Worsnop et al. 1993), and may have a role in the development of the frozen particles that have also been observed (Tabazadeh et al. 1995). In addition, subgrid-scale temperature perturbations, such as caused by stratospheric gravity waves, may act to provide additional intermittent chemical processing or induce freezing of PSC particles (Meilinger et al. 1995). Further, the rates of some key reactions on heterogeneous surfaces are still quite uncertain on most of the hypothesized particle types.

The effect of binary sulphate aerosols produced from volcanic eruptions may also be significant (Knight et al. 1998). This may have led to an underprediction in the area of the ozone hole and might also contribute to mid-latitude ozone loss. These processes will be incorporated in a future version of the model.

(c) **Model temperature correction**

In order to simulate global ozone amounts accurately, the temperatures used in the heterogeneous calculations were increased by 5 K. This can be justified by considering the model temperature bias relative to observations and the use of thermodynamic equilibrium in the PSC scheme. In the global mean the temperature bias is about −3 K at 46 hPa, relative to Stratospheric Sounding Unit data. In addition, some authors assume a nucleation barrier of about 3 K for NAT and 2 K for ice (for example, Steil et al. (1998), and the NASA Goddard model—IPCC (1999, chapter 4)). The two processes combine to give an effect of about 5–6 K. However, following the work of Lefèvre et al. (1998), we will consider using ternary-solution microphysics. Adjustments to the orographic gravity-wave drag scheme of the model (Jackson et al. personal communication) to increase temperatures in the lower stratosphere may also allow the temperature correction to be eliminated or reduced.

(d) **Comparison with previous work**

The model results confirm previous results (Shindell et al. 1998) in that increases in greenhouse gases in the current atmosphere are likely to lead initially to increased ozone depletion, even though some chlorine reduction is anticipated. However, the ozone perturbations calculated here are much smaller than the magnitude predicted by Shindell et al. (1998). While they use a somewhat simplified chemistry scheme, a major impact on the model results is the predicted frequency of stratospheric warmings which, as noted in subsection 8(a), can be highly significant. Other model results presented in chapter 12 of WMO/UNEP (1999) show insignificant differences between the results for the 1990s and 2010s. However, the ECHAM3 model (Steil et al. 1998; Dameris et al. 1998) includes tropospheric chemistry and, despite lower ozone in high latitudes, they simulate higher total ozone in 2015 than in 1991 (WMO/UNEP 1999, chapter 12). Apart from these general characteristics, direct comparison between the three-dimensional
model results is difficult due to differing amounts of halogens specified, as well as other important details. Consequently, further more definitive calculations need to be done to resolve some of the current discrepancies.

9. CONCLUSION

In this work we have simulated the past and future state of the ozone layer using one-year integrations of a three-dimensional coupled chemistry–climate model. The results for 1980 and 1995 are in reasonable agreement with observations while the simulations for future years indicate increased ozone depletion due to the increased levels of greenhouse gases. The model begins to indicate the first signs of recovery of total ozone in about 2010, although recovery in other regions, such as the Arctic, may be delayed a further 5 years. As anticipated by Hofmann and co-workers (Hofmann et al. 1995; Hofmann 1996; Hofmann et al. 1997) the model suggests that Antarctica will be the best region to look for this recovery. In particular, the model results for 2014 suggest that, at least occasionally over the next few decades, the ozone hole could become much less deep and approach the depth last observed in about 1980. Since halogen levels are expected to be quite high in 2014, this partial recovery of the Antarctic ozone hole might be a temporary situation and predictions of the evolution of the ozone hole are likely to be very challenging. Also, the results suggest that total ozone will continue to decrease in the tropics, although the model decrease is slightly larger than currently observed. The impact on surface UV is predicted to be relatively small for a large part of the earth. However, in the tropics, where UV is high because ozone and solar zenith angles are lower than in mid and high latitudes, clear-sky UV is predicted to increase up to 8% relative to 1980. Over Antarctica, the effects are likely to be even more serious, as the predicted increased stability of the ozone hole would enhance clear-sky UV by over 35% in the zonal mean in early summer by about the year 2005. Local enhancements could lead to more than a factor of two in UV variability around the latitude circle. The results also have implications for climate change in that for the period 1980–2015 the coupled chemistry–climate simulation exhibited a cooling rate in the lower stratosphere of twice that found in a similar model simulation without chemistry.

Three-dimensional coupled chemistry–climate models are still in their relative infancy and ideally one would wish to perform continuous multi-decadal simulations with changing GHGs. Now that the model has been tested for one-year snapshots we are in a position to commence this, after first upgrading the model microphysics and verifying the long-term stability of the model. Finally, the conclusions reached in this work are based on current predicted trends in halogen-containing molecules. Future monitoring of these molecules is therefore essential, given the fragile nature of the ozone layer.

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APPENDIX

Rescaling the conservative horizontal tracer

Tracer-1, which is initialized as the potential-vorticity equivalent latitude on a model surface, is advected by the model but is also modified once every model day. This
prevents the effects of the unrealistic diffusion that results from the use of the model tracer-transport scheme. In the recalculation procedure, the new value of Tracer-1, \( \tilde{\chi}(\lambda_0, \phi_0) \), at the grid point with longitude \( \lambda_0 \) and latitude \( \phi_0 \), is set equal to the latitude which encloses the same area on the globe as the contour of the existing Tracer-1, \( \chi(\lambda_0, \phi_0) \). Summation over all the grid points on the model surface within the contour gives this area which, expressed in units of the radius of the earth squared, is

\[
A(\lambda_0, \phi_0) = \sum_{\chi(\lambda, \phi) \geq \chi(\lambda_0, \phi_0)} a(\phi) \Delta \lambda, \tag{A.1}
\]

where \( \lambda \) is longitude, \( \phi \) latitude, \( \Delta \lambda \) the longitudinal grid spacing and \( a(\phi) \) is defined as

\[
a(\phi) = \begin{cases} 
1 - \sin(\phi - \Delta \phi/2) & \text{for } \phi = \frac{\pi}{2} \\
1 + \sin(\phi + \Delta \phi/2) & \text{for } \phi = -\frac{\pi}{2} 
\end{cases} \tag{A.3}
\]

with \( \Delta \phi \) the latitudinal grid spacing. The new value of Tracer-1 is then obtained by inverting the expression for the area within the latitude circle,

\[
\tilde{\chi}(\lambda_0, \phi_0) = \sin^{-1} \left( 1 - \frac{A(\lambda_0, \phi_0)}{2\pi} \right). \tag{A.4}
\]

In practice, this is achieved most efficiently by placing the grid-point values of \( \chi \) into descending order, resulting in the transformation \( \chi(\lambda, \phi) \rightarrow \chi_i \) where the \( i \)th value in the sequence was originally at coordinates \((\lambda_i, \phi_i)\). The summation Eq. (A.1) then becomes

\[
A_i = \sum_{j=1}^{i} a(\phi_j) \Delta \lambda, \tag{A.5}
\]

and the new value of Tracer-1 follows by

\[
\tilde{\chi}(\lambda_i, \phi_i) = \sin^{-1} \left( 1 - \frac{A_i}{2\pi} \right). \tag{A.6}
\]

The corrected model tracer fields are more closely conserved and remain related to the dynamical fields on each model level over long integrations. Strictly, it should be applied to Tracer-1 values on a Lagrangian surface (and clearly model surfaces diverge from such surfaces), but the differences are unlikely to introduce substantial inaccuracies to the results over a long timescale.

REFERENCES


Butchart, N, Austin, J., Gallani, M., Knight, J. R. and Scaife, A., A. 2000 The response of the stratospheric climate to projected changes in the concentrations of well-mixed greenhouse gases from 1992 to 2051. *J. Climate*, in press


McKinlay, A. and Difley, B. L.


Middlebrook, A. M., Koehler, B. G., McNeill, L. S. and Tolbert, M. A.


Mitchell, J. F. B. and Johns, T. C.

1995 Climate response to increasing levels of greenhouse gases and sulphate aerosols. Nature, 376, 501–504


1996 Decline in the tropospheric abundance of halogen from halocarbons: Implications for stratospheric ozone depletion. Science, 272, 1318–1322


Plumb, R. A. and Ko, M. K. W.


Oltmans, S. J. and Hofmann, D. J.


Ramawamy, V., Schwarzkopf, M. D. and Randel, W. J.


Rind, D., Shindell, D., Lonergan, P. and Balachandran, N. K.

1997 Mid-winter start to Antarctic ozone depletion: Evidence from observations and models. Science, 278, 93–96

Roscoe, H. K., Jones, A. E., Lee, A. M.


Shindell, D. T., Rind, D. and Lonergan, P.


Solomon, S., Portmann, R. W., Garcia, R. R., Thomason, L. W., Poole, L. R. and McCormick, M. P.

1996 SORG

Steig, B., Dameris, M., Bruhl, C., Crutzen, P. J., Grewe, V., Ponater, M. and Sauser, R.

1993 Monitoring stratospheric ozone from space. Pp. 319–346 in The role of the stratosphere in climate change. NATO Advanced Study Institute Series I, Environmental change, 8, Springer-Verlag

Stolarski, R. S.


Swinbank, R. and O’Neill, A.


Tabazadeh, A., Toon, O. B. and Hamill, P.


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WMO/UNEP


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Worsnop, D. R., Fox, L. E., Zahniser, M. S. and Wofsy, S. C.