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Chemical ozone loss in a chemistry-climate model from 1960 to 1999

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[1] In the recent WMO assessment of ozone depletion, the minimum ozone column is used to assess the evolution of the polar ozone layer simulated in several chemistry-climate models (CCMs). The ozone column may be strongly influenced by changes in transport and is therefore not well-suited to identify changes in chemistry. The quantification of chemical ozone depletion can be achieved with tracer-tracer correlations (TRAC). For forty Antarctic winters (1960–1999), we present the seasonal chemical depletion simulated with the ECHAM4.L39(DLR)/CHEM model. Analyzing methane–ozone correlations, we find a mean chemical ozone loss of 80 ± 10 DU during the 1990s, with a maximum of 94 DU. Compared to ozone loss deduced from HALOE measurements the model underestimates chemical loss by 37%. The average multidecadal trend in loss from 1960 to 1999 is 17 ± 3 DU per decade. The largest contribution to this trend comes from the 62 ± 11 DU ozone loss increase between the 1970s and 1990s.

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1. Introduction

[2] To simulate the past and future development of the ozone layer is one of the major tasks for chemistry-climate models (CCMs). Mechanistic models have been used since the early 1990s [e.g. *Granier and Brasseur*, 1991]. Fully coupled three-dimensional CCMs emerged during the mid-1990s [e.g. *Déqué and Piedelievre*, 1995; *Dameris et al.*, 1998] and are still in development. In spite of known deficiencies, CCM simulations are the only way to achieve a self-consistent view of atmospheric state in the past and the future, and to examine its interaction with climate change. The performance of CCMs has been compared by *Austin et al.* [2003] and V. Eyring et al. (Assessment of temperature, trace species and ozone in chemistry-climate simulations of the recent past, submitted to *Journal of Geophysical Research*, 2006, hereinafter referred to as Eyring et al., submitted manuscript, 2006). An assessment of the skill of CCMs is provided by the process-oriented CCM validation (CCMVal) [*Eyring et al.*, 2005] activity supported by the World Climate Research Programme (WCRP). Amongst the processes to be investigated is the representation of polar chemical O_3 loss.

[3] The springtime minimum O_3 column has been used almost exclusively to assess long-term polar O_3 changes in models [*Austin et al.*, 2003; *World Meteorological Organization* (*WMO*), 2003; *Intergovernmental Panel on Climate Change* (*IPCC*), 2005]. This minimum column is a combined result of transport and chemistry. Changes in this quantity therefore cannot be separated from changes due to chemistry alone. To evaluate the effect of chlorofluorocarbon emission reductions on polar O_3 it is the change in the chemical regime under concurrent climate change which is most interesting. By employing a seasonal proxy for chemically undisturbed polar O_3, the amount of chemical O_3 depletion can be calculated [*Chipperfield et al.*, 1993; *Manney et al.*, 1995; *Goutail et al.*, 1999]. While such a proxy is routinely used in chemistry transport simulations of the past decade, it is not yet available for the majority of CCMs. The tracer-tracer correlation technique (TRAC) obviates this problem by using an early winter relationship between a chemically long-lived tracer and O_3 to derive a chemically unperturbed O_3 proxy at any later time during polar winter and spring [*Proffitt et al.*, 1990]. Recently, *Lemmen et al.* [2006] demonstrated that for the ECHAM4.DLR(L39)/CHEM (abbreviated E39/C) model, tracer-tracer correlations can be employed to identify chemical O_3 loss even on CCM data. TRAC is applied here on the forty-year E39/C simulation 1960–1999 which is based on the REF1 scenario for the upcoming WMO assessment of O_3 depletion (expected to be published in 2007).

2. Data and Analysis

[4] The ECHAM4.DLR(L39)/CHEM coupled chemistry-climate model system is described in detail by *Steil* [1998] and *Steil et al.* [1998]; E39/C is run at T30 horizontal resolution and has 39 vertical layers up to 10 hPa. Chemical tracers are transported by a semi-Lagrangian scheme [*Williamson and Rasch*, 1994]. Chemical changes to radiatively active gases provide feedback to the dynamical scheme via their heating rates. The 40-year simulation from 1960 to 1999 is presented by *Dameris et al.* [2005]. Sea surface temperatures were provided according to HadISST1 and HadCM3; *IPCC* [2001] A1b and *WMO* [2003] CFC and greenhouse gas emission scenarios were assumed. The model was nudged to observed equatorial winds to reproduce the quasi-biennial oscillation; cloud radiative feedback and major volcanic eruptions are considered. The solar cycle representation in the model is investigated by *Dameris et al.* [2006] and shown to delay the expected onset of ozone recovery in the model.

[5] Tracer-tracer relations in the vortex were first employed by *Roach* [1962] to distinguish air masses; later, they were used to quantify chemical O_3 loss [*Proffitt et al.*,

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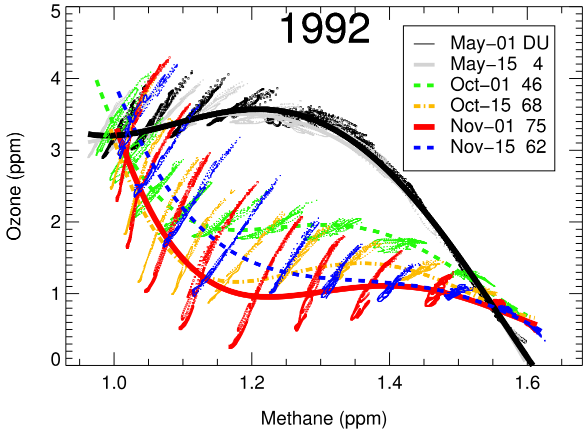

Figure 1. Tracer-tracer correlation for the Antarctic winter 1992 (ϕ poleward 70°S) for first and 15th of each month May, October, and November. Column loss (DU) is integrated over altitude range 380–640 K where chemical loss is evident. Though in some winters of the 1990s CCM O_3 is completely removed from a single altitude level, O_3 is not depleted fully over a broad altitude range, in contrast to observations.

1990]. In the absence of mixing, correlations between a long-lived tracer and ozone provide a quantitative measure of chemical O_3 change [*Müller et al.*, 2001, 2005]. TRAC has been successfully used to quantify chemical O_3 loss for the past decade of Arctic [*Tilmes et al.*, 2004] and Antarctic winters (S. Tilmes et al., Chemical ozone loss in the Arctic and Antarctic stratosphere between 1992 and 2005, submitted to *Geophysical Research Letters*, 2006, hereinafter referred to as Tilmes et al., submitted manuscript, 2006a). The performance of TRAC was investigated by a number of studies for the 1999/2000 Arctic winter [e.g. *Salawitch et al.*, 2002; *Ray et al.*, 2002]. A comparison of TRAC to other techniques by *Harris et al.* [2002] showed general agreement of TRAC-derived O_3 loss with O_3 loss derived from Lagrangian trajectory [*Manney et al.*, 1995], vortex average [*Knudsen et al.*, 1998], SAOZ [*Goutail et al.*, 1999] and Match [*von der Gathen et al.*, 1995] calculations.

[6] Mixing across the polar vortex edge impacts any empirical method to determine chemical O_3 loss and was considered problematic for TRAC [*Michelsen et al.*, 1998; *Plumb et al.*, 2000], especially in the more dynamically active Arctic. On must be cautious applying TRAC to data from CCMs which may exhibit high numerical diffusivity. *Sankey and Shepherd* [2003] find that the Canadian Middle Atmosphere Model's northern polar vortex has "virtually no transport barrier" which would be necessary for TRAC to function. For E39/C, *Lemmen et al.* [2006] provide evidence that TRAC yields reasonable, if underestimated, values for cold future Arctic winter chemical O_3 loss. They further demonstrate that the error introduced by mixing on TRAC ozone loss estimates is minimized at low altitudes such that O_3 column calculations are less severely impacted than calculations of local O_3 loss at greater altitudes (above ≈500 K).

[7] The Antarctic polar vortex is better isolated and more stable than the Northern polar vortex and thus better suited to TRAC analysis [*Sankey and Shepherd*, 2003]. Like in all other CCMs, the southern hemisphere vortex in E39/C is more stable and persists longer than in observations (Eyring et al., submitted manuscript, 2006). For all 40 model years, we determine the seasonal chemical O_3 loss between May 1 and November 1 of each year. In May during all years, a stable vortex has formed and the relationship between O_3 and CH_4 is compact. The vortex persists at least until November 1 in all years.

[8] For spatial averaging the area poleward of 70°S equivalent latitude (ϕ) was chosen, corresponding to the mean and median May 1 vortex edge at 475 K of the 40 year time series. The vortex edge was determined as the equivalent latitude, where the gradient of potential vorticity with respect to ϕ was largest and near the maximum zonal wind [*Nash et al.*, 1996]. We did not correct for a linear trend of 1.3° (not significant at 95% confidence level) found in the vortex edge determination.

[9] Vertically, isentropic levels between 380 and 640 K were considered (≈16–27 km or 100–20 hPa). The 4th order polynomial fit through the median values of CH_4 and O_3 at each isentropic level represents the compact tracer relationship in each spring well (Figure 1). The choice of the median value on isentropes removes the influence of outliers which are due to the low upper domain boundary of the model [*Lemmen et al.*, 2006], and represents the overall vortex better than the mean.

3. Results

[10] Histograms of the southern hemispheric probability density distribution of individual pairs of CH_4 and O_3 mixing ratios on May 1 (not shown) reveal two distinct relationships with high occurrence of tracer pairs: (1) a linear relationship represents subtropical and mid-latitude values; (2) a convex polar relationship represents high mid-latitude and polar tracer values [e.g. *Proffitt et al.*, 2003; S. Tilmes et al., Development of tracer relations and chemical ozone loss during the setup phase of the polar vortex, submitted to *Journal of Geophysical Research*, 2006]. The existence of these distinct relationships is evidence for a transport barrier in the atmosphere. Between these two distinct relationships a continuous range of tracer-tracer pairs is seen in the data which shows that the transport barrier does not completely isolate the tropical, mid-latitude and polar air masses in the model [*Plumb*, 2002].

[11] For equivalent latitudes poleward of 70°S, compact vortex relationships are found for each of the 40 model years. The degree of compactness can be categorized from the root-mean-square (rms) error of a least-squares (4th order) polynomial fit to the vortex data. The rms range for the 40 winters is 110–280 parts per billion by volume (ppb). The most compact relationships (rms < 150 ppb) are found in winters 1980, 1991–93, and 1995. The least compact (rms > 250 ppb) relationships occur in winters 1961, 1964, 1971, 1976–77, and 1985–86; all winters of the 1990s have very compact early winter relationships with rms < 220 ppb. The reference relationship derived from the compact vortex data in early winter can be used to calculate a chemically undisturbed O_3 proxy from the CH_4 field.

[12] The vortex CH_4-O_3 relationship is compact for all model years in fall. Larger variability within the vortex is present in austral spring, evident as large rms values. The increase of variability within each isentropic layer from fall

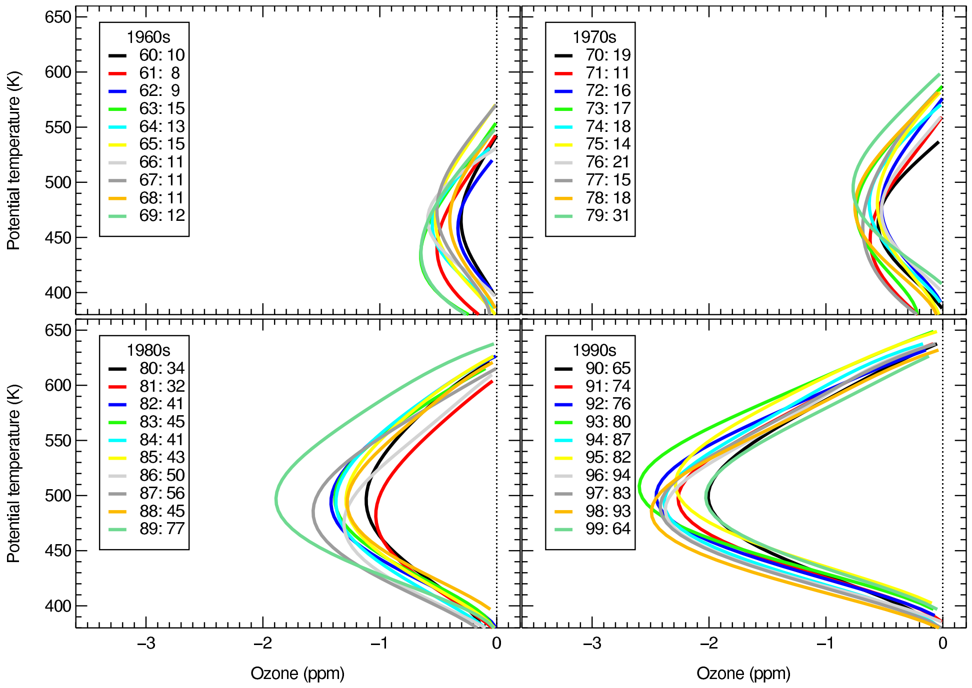

Figure 2. Average Antarctic (ϕ poleward of 70°S) chemical O_3 loss profiles for each decade 1960–1999, individual years indicated by color. May to November chemical O_3 loss is analyzed with CH_4-O_3 tracer correlations, only the part of the O_3 column where chemical loss is evident is evaluated, numbers given for integrated O_3 loss in the legend for each year are in Dobson units (DU).

to spring is a result of the counteraction of chemical O_3 depletion and mixing with less depleted mid-latitude air. While not representative for the entire vortex, the lower domain boundary of tracer values in Figure 1 may be thought of as the maximum action of chemistry on vortex O_3. Evidence, but not proof, that this domain boundary is unaffected by mixing is given by the fact that for several winters of the 1990s, vortex O_3 is completely lost from some model levels: for example, the minimum O_3 mixing ratio between 400–450 K in 1996 and 1998 is less than 50 ppb.

[13] For each of the 40 model years, the chemically undisturbed O_3 proxy is calculated from the CH_4 field on November 1. Chemical O_3 loss is evaluated as the difference between the simulated O_3 field and the correlation-derived O_3 proxy. A vortex-average loss profile is derived by averaging vortex chemical loss on each model level (Figure 2). The decadal intensification of chemical O_3 loss from the 1960s to the 1990s is evident in the increasing magnitude of maximum loss and the extension of the vertical range of chemical O_3 loss. In the 1960s, a maximum loss of 0.65 ppm occurred; the top altitude of chemical loss did not exceed 570 K. In subsequent decades, the model simulates the largest chemical loss around 500 K. In the 1970s, maximum chemical loss is 0.8 ppm; in the 1980s, up to 1.9 ppm O_3 are lost—averaged over the vortex area at 500 K. In the 1990s, chemical O_3 loss is evident over the entire considered altitude range 340–640 K; averaged maximum loss of 2.6 ppm occurs at 500 K in the year 1993.

[14] The temporal evolution of chemical O_3 loss from 1960 to 1999 is shown in Figure 3 (top). During the 1960s until the mid-1970s, very little chemical loss occurred. From the end of the 1970 to the 1990s a strong increase is seen in the chemical loss column. Largest loss simulated with E39/C and deduced with TRAC is 94 DU in winter 1996. Compared to chemical loss deduced from HALOE satellite measurements (Tilmes et al., submitted manuscript, 2006a) the model underestimates O_3 loss by 37% (82 DU vs. 131 DU, 1992–1997). The HALOE vortex core calcu-

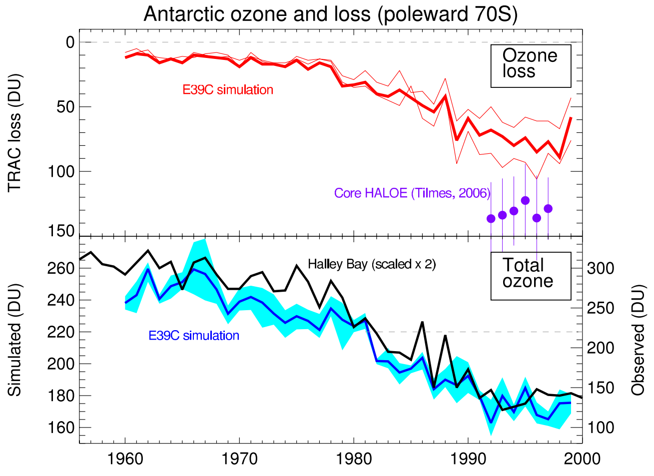

Figure 3. Antarctic O_3 and chemical O_3 loss 1960–1999 in E39/C. (top) TRAC-derived chemical O_3 loss within ϕ = 70°S contour from E39/C simulation (solid line) and from satellite observations (dots). (bottom) Monthly mean October total O_3. The range of an ensemble of three E39/C simulations from 1960 to 1999 (cyan shading) and their mean value (blue line) are shown. Halley Bay Dobson spectrometer observations (black) are shown scaled to the simulated data, absolute values shown on right axis.

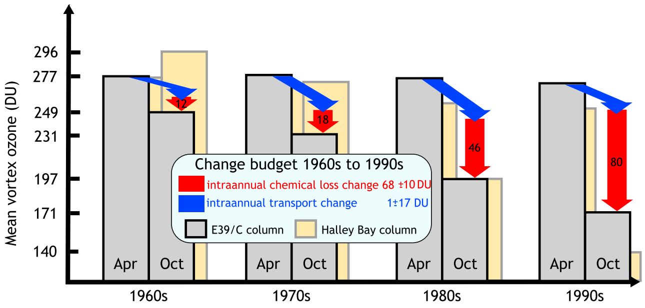

Figure 4. Decadally averaged total O_3 in April and October, poleward of 70°S. Model results shown as grey bars, Halley Bay observations as beige bars. Red arrows indicate the magnitude of chemical O_3 loss deduced from the E39/C simulation. Blue arrows show the remaining difference between intra-annual total O_3 change and chemical change.

lations are shown in Figure 3 as purple dots. Largest HALOE-observed loss is 137 ± 28 DU for the year 1992; bars show the reported uncertainty in the reference relation (on average ±28 DU).

[15] Figure 3 (bottom) shows the evolution of October monthly mean total O_3 from 1960 to 1999 in E39/C (poleward of 70°S) and from 1956 to 2000 above Halley Bay (75.5°S, data updated from *Jones and Shanklin* [1995] and shown scaled by factor 2, aligned with E39/C at 220 DU). Both time series agree qualitatively: The 220 DU threshold for an "ozone hole" [*WMO*, 1999] is transgressed by both series after the year 1980; minimum values occur during the 1990s. For most years prior to 1992, both time series exhibit cotemporal annual variation, most pronounced in common positive anomalies in years 1962, 67, 71, 74, 78, 81, 86, 88, and 1990.

[16] Quantitatively, the model reproduces only half of the observed total decline of fall polar O_3: The pre-ozone hole average October mean column (1964–1980) in observations is 281 DU versus 237 DU in the simulation. The 1990s average October mean column in observations is 141 DU, compared to 175 DU in the model. From the mean April column values in both time series (pre-1980: E39/C: 279 DU, Halley Bay: 270, several years without measurements), a systematic offset of the model data of about 10 DU can be estimated; this April offset increases to 25 DU (E39/C: 274 DU, Halley Bay 253 DU) during the 1990s.

[17] The decline from the pre-ozone hole era to the 1990s is −140 DU observed and −62 DU simulated. This underestimation of the post-1980 trend by E39/C by a factor of 2 was reported by Eyring et al. (submitted manuscript, 2006) in their model intercomparison study. In spite of the cold pole bias (w.r.t. UK MetOffice analyses) of up to −20 K in austral winter in E39/C, not enough O_3 is chemically destroyed due to underestimated total chlorine (Cl_y) and the neglect of Br chemistry in the lower stratosphere. Eyring et al. (submitted manuscript, 2006, Figure 12) attribute the underestimation of Cl_y in the polar lower stratosphere (Cl_y > 1.7 ppb) to insufficient transport from the upper model domain boundary (where Cl_y is prescribed in E39/C) to 100–70 hPa.

[18] In Figure 4, we compare decadally averaged intra-annual total O_3 change and chemical O_3 loss. Compared to the Halley Bay time series (beige color), additional deficiencies of the model (grey bars) regarding transport and chemistry become evident. (1) The model simulates a constant April vortex column throughout the four decades, whereas in the Halley Bay measurements as small long-term decline (−7 DU per decade) is present. (2) Dynamical resupply should lead to an increase of the O_3 column from winter to spring in a chemically unperturbed regime like the 1960s. In the 1960s model data instead, a total O_3 decrease of 28 DU is seen, of which 12 ± 2.5 DU are identified as caused by chemistry (red arrow). Whereas in the measurements, a resupply of at least 20 DU occurs, in the model, a transport-related decrease on the order of 16 DU is seen (blue arrow).

[19] Chemical O_3 loss increases with time, the difference between the 1990s and 1960s E39/C mean chemical O_3 loss is 68 ± 10 DU, or 23 DU per decade. This trend is larger from the 1970s to 1980s (28 ± 14 DU) and from the 1980s to the 1990s (34 ± 11 DU). By calculating the difference of the total intra-annual ozone change and the chemical depletion, one obtains a remaining term which contains transport-related changes and errors in the ozone loss calculation. While large errors accumulate in this deduced quantity, shown as blue arrows in Figure 4, there is no significant decadal change.

4. Summary and Conclusion

[20] From the forty year E39/C REF1 simulation 1960–1999, the chemical contribution to O_3 change was successfully extracted using the tracer-tracer correlation technique. A multidecadal trend in chemical O_3 loss of 17 DU per decade was found. The model simulates large chemical O_3 losses for the 1990s, but only 63% of the chemical loss found from observational data. We attribute this underestimation largely to the known insufficient chlorine loading in the model rather than to a systematic underestimation by the chosen methodology. Unlike observations, the model does not simulate the complete removal of O_3 over a broad altitude range: the CCM is expected to react more sensitively to the expected Cl_y decrease and thus to predict a predated early sign of recovery. A budget of total and chemical O_3 change for each of the four decades shows that the intra-annual winter to spring O_3 resupply in the polar vortex is not well simulated.

[21] As a measure of Antarctic O_3 depletion, *Bodeker et al.* [2005] argue that the minimum O_3 column is not the preferred quantity. However, it is readily available from CCMs and has thus been the focus of past and upcoming assessments of the evolution of the O_3 layer in CCMs. We show here that one can obtain additional information on the chemical contribution to O_3 change in a CCM with equally easily accessible data from CCMs. To our knowledge, chemical O_3 loss has so far been analysed from E39/C time slice experiments [*Lemmen*, 2005] and from the WACCM CCM (S. Tilmes et al., Evaluation of heterogeneous processes in the polar lower stratosphere in WACCM3, manuscript in preparation, 2006). We encourage other CCM groups to participate in this effort to extract the

chemical signal of O_3 loss from their current and archived experiments.

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