



Comparison of measured and modeled ozone above McMurdo Station, Antarctica, 1989–2003, during austral winter/spring

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Received 30 August 2006; revised 21 June 2007; accepted 3 July 2007; published 12 October 2007.

[1] Fifteen years of ozonesonde measurements at McMurdo Station, Antarctica (78°S), are used to test the ability of the SLIMCAT off-line 3-D chemical transport model to reproduce Antarctic stratospheric ozone in the period of August–October over many annual cycles. Two versions of SLIMCAT, both previously used in Arctic studies, are used in a detailed quantitative comparison for total column ozone and ozone mixing ratios (O_3MR) at the vertical resolution of SLIMCAT. The newer model run, EC, forced by ERA-40 ECMWF meteorological (re)analyses, uses a general circulation model (GCM) radiation scheme to derive vertical transport and has improved chemistry and denitrification. The older run, UK, uses UKMO analyses and the MIDRAD scheme. Run EC shows good agreement with measured total column ozone (within $\pm 10\%$). Run UK has similar agreement except during the annual ozone depletion period when run UK overestimates total column ozone by $\sim 20\%$ in many years. Linear regression of O_3MR shows run EC is in excellent agreement with measurements, $R^2 = 0.89$. Run UK does not agree as well at higher altitudes in late winter. Overall, the newer version of SLIMCAT, with improved vertical velocity due to the use of the GCM radiation scheme and improved vertical and temporal resolution due to the use of ERA-40 ECMWF (re) analyses, does well in reproducing variations in Antarctic ozone concentrations over many annual cycles, indicating SLIMCAT can make useful contributions to chemistry climate models (CCM).

Citation: Mercer, J. L., C. Kröger, B. Nardi, B. J. Johnson, M. P. Chipperfield, S. W. Wood, S. E. Nichol, M. L. Santee, and T. Deshler (2007), Comparison of measured and modeled ozone above McMurdo Station, Antarctica, 1989–2003, during austral winter/spring, *J. Geophys. Res.*, 112, D19307, doi:10.1029/2006JD007982.

1. Introduction

[2] Measurements are needed for characterizing the past and present state of the atmosphere, while models represent our quantitative understanding of atmospheric processes. Long-term self-consistent data sets are essential for detecting and analyzing trends and providing robust and critical measurements for comparison with models. As photochemical box models, general circulation models (GCM), and chemical transport models (CTM) have been refined, so

have the tests of these models against a variety of observations [e.g., Grose *et al.*, 1987; Rood *et al.*, 1989; Kaye *et al.*, 1990; Lefèvre *et al.*, 1994; Chipperfield *et al.*, 1994; Eyring *et al.*, 2005]. Early comparisons between measured ozone and that estimated by CTMs involved short timescales, and mainly concentrated on the Arctic.

[3] A 1994/1995 study in the Arctic compared total column ozone from measurements taken at Sodankylä (67°N, 26°E) and at Zhigansk (66°N, 123°E) with total column ozone calculated by the REPROBUS model (of CNRM-Meteo-France [Lefèvre *et al.*, 1994; Goutail *et al.*, 1999]). The model was forced by the European Centre for Medium-range Weather Forecasts (ECMWF) analyses and captured the day-to-day variations in ozone quite well, but overestimated ozone mixing ratio profiles by 5% [Goutail *et al.*, 1999]. The overestimation in the north was most prominent after 1 January of each year and the authors suggested that the overestimation could be because the ClO/BrO chemistry was not adequately accounted for in the model. Another model, the MIMOSA-CHIM [Lefèvre *et al.*, 1994; Hauchecorne *et al.*, 2002; Marchand *et al.*, 2003], was compared to Arctic ozonesonde measurements of total column ozone in 1999/2000 and was found to underestimate ozone mixing ratios at 435 K and 475 K during the onset of

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annual ozone depletion [Marchand *et al.*, 2003]. More recently, the MIMOSA-CHIM was used to simulate ozone loss over three Arctic and three Antarctic winters with an accuracy of about 20–30%. It was concluded that the model requires improvements, particularly in the area of vertical transport [Tripathi *et al.*, 2006]. Other comparisons with measurements in the Arctic [Sinnhuber *et al.*, 2000; Kilbane-Dawe *et al.*, 2001] indicated that the SLIMCAT model forced by the United Kingdom Met Office (UKMO) analyses overestimated total column ozone, results which are similar to the results of Goutail *et al.* [1999] where the REPROBUS model was used.

[4] Photochemical box models have also been used to calculate ozone loss rates in the Arctic. One such model was compared with data from the Improved Limb Atmospheric Spectrometer (ILAS) and with ozonesondes flown over the Arctic. The model was found to calculate ozone loss rates reasonably well in general, though the study was preliminary and a detailed statistical comparison was not completed [Kagawa and Hayashida, 2003].

[5] In the Antarctic, a long-term study compared SLIMCAT estimates of total column ozone with Dobson spectrophotometer measurements at Halley Bay (76°S) from late 1991 through early 1997 [Chipperfield, 1999]. The SLIMCAT model was forced using the UKMO analyses and reproduced the overall trend of annual ozone fluctuations quite well; however, it overestimated total column ozone (by ~20%) during the austral spring when the rate of ozone depletion is at its greatest, and during austral summer (by ~17%) when the polar ozone layer is replenished with ozone-rich air from the midlatitudes. Chipperfield [1999] suggested that the overestimation could be due to a modeled poleward/downward transport of O₃ during the summer that was too strong, or to a chemical sink in the summer polar lower stratosphere that was too slow, or both. Lee *et al.* [2000] compared the SLIMCAT model with ozonesonde measurements over Antarctica during 1996. While the results showed generally good agreement between the model and measurements, it was noted that the model systematically overestimated ozone during winter [Lee *et al.*, 2000].

[6] Recently, a 40-year run of the ECHAM4.L39(DLR)/CHEM [Steil *et al.*, 1998] model showed an underestimation of ozone loss by 37% over Antarctica, which was attributed to insufficient chlorine loading [Lemmen *et al.*, 2006].

[7] The sources of the observed differences between models and measurements in these studies are generally thought to be due to either inadequate model chemistry or inadequate model transport, or both. A study over Antarctica compared POAM III satellite data with a photochemical box model driven by different types of trajectories and found trajectory errors to be the largest source of uncertainty when comparing models with measurements [Hoppel *et al.*, 2005]. The ECMWF analysis was found to be more accurate than UKMO or NCEP when used to drive trajectories for the CTM's determination of ozone loss rates [Hoppel *et al.*, 2005]. The study experimented with a range of values for ClOOCl cross sections and ClOx and BrOx concentrations and found that the most accurate results were found with ECMWF winds when using minimal acceptable amounts of ClOx and BrOx and the ClOOCl cross sections

of JPL 2002, but for UKMO or NCEP winds ClOx and BrOx had to be significantly increased and the higher ClOOCl cross sections of Burkholder *et al.* [1990] had to be used [Hoppel *et al.*, 2005]. Estimates of ozone mixing ratios by the box model Chemical Lagrangian Model of the Stratosphere (CLaMS) [McKenna *et al.*, 2002a, 2002b] were compared with Improved Limb Atmospheric Spectrometer (ILAS-II) measurements over Antarctica and showed generally good agreement, however, the ILAS-II observations have large day-to-day variations in ozone mixing ratios due to differences in air masses [Tilmes *et al.*, 2006]. A study comparing tracers estimated by the UMSLIMCAT model, a CCM that bases its chemistry on the SLIMCAT model [Tian and Chipperfield, 2005], with HALOE measurements found good agreement for methane, HCl, mean air age and propagation of water vapor across the tropopause [Eyring *et al.*, 2006].

[8] Most of the above studies comparing measured and modeled ozone have contributed to the notion that models underestimate polar chemical ozone loss. However, an updated version of SLIMCAT, forced with the ECMWF ERA-40 reanalyses (operational analyses after 2000), and with improved vertical transport and chemistry, has proven to reproduce well observed ozone losses in the Arctic, including profile shapes and year-to-year variability [Chipperfield *et al.*, 2005]. Comparisons of this updated version of SLIMCAT with balloon-borne ozonesonde measurements over the Arctic do not show the same underestimation of loss that the earlier versions have shown [Feng *et al.*, 2005].

[9] Given this improvement in our ability to model chemical loss over the Arctic, it is useful to compare the same global model runs with observations in the Antarctic as a further test of our understanding of polar chemistry and transport. Here, we present a detailed comparison of 15 years of balloon-borne ozonesonde measurements above McMurdo Station, Antarctica (78°S) with old and new runs of the SLIMCAT CTM. The aim is to quantitatively assess the agreement between model estimations of total column ozone and O₃MR and in situ measurements of these quantities in the Antarctic at 78°S. Two versions of the model are used; an old run [Lee *et al.*, 2000; Chipperfield, 1999] which is forced by UKMO analyses [Swinbank and O'Neill, 1994], and the newer version used by Chipperfield *et al.* [2005]. These are standard runs of the SLIMCAT model (runs 198 and 323) and any changes between the two runs were made to improve the model overall, thus there were no changes made specific to this study. By using a long-term record of ozone profile observations we are able to test the ability of the model to capture, in two different multiyear runs, detailed changes in polar ozone and its interannual variability.

2. Measurements and Model

2.1. Balloon-Borne Ozonesonde Measurements

[10] Ozonesondes have been routinely flown from McMurdo Station (77.8°S, 166.7°E) between ~21 August and 1 November of each year since 1986. In addition, austral winter flights were made in 1994 and 2003. The sondes are flown with Vaisala radiosondes to measure ambient temperature and pressure. Balloon flights typically reach altitudes between 30 and 35 km. Total column ozone

values (calculated using constant mixing ratio extrapolation) have been routinely compared to Dobson spectrophotometer measurements taken at Arrival Heights, 1 km from McMurdo Station, and to satellite measurements (TOMS, EP-TOMS and SBUV-2) and typically show agreement to within $\pm 5\%$ [Deshler and Hofmann, 1991; Johnson *et al.*, 1994, 1995; Nardi *et al.*, 1997, 1999; Kröger *et al.*, 2003]. Exceptions occur when ozone gradients are high near the polar vortex edge [Deshler *et al.*, 1990]. Ozonesonde measurements are considered to have a precision of $\pm 5\%$ [Komhyr *et al.*, 1995].

[11] The electrochemical concentration cell ozonesondes for McMurdo measurements have always been prepared using standard procedures with a 1% KI buffered solution, except in 1999, when 0.5% KI buffered solution was used. Each ozonesonde pump is individually calibrated in the laboratory for efficiency between 100 and 5 mbar [Johnson *et al.*, 2002]. While the use of 1.0% KI cathode sensing solution has been maintained at McMurdo, the ozonesonde manufacturer was switched from Science Pump (SP) to ENSCI (ES) in the mid 1990s. Several years later, in 1996, ES recommended that 0.5% KI cathode solution be used instead of 1.0% KI. This recommendation came after a preliminary analysis of laboratory tests suggested that ES sondes with 1.0% KI overestimated ozone [Smit *et al.*, 2007]. Since then, additional laboratory studies [Smit *et al.*, 2007] and field comparisons (T. Deshler *et al.*, Field test of ECC-ozonesondes from different manufacturers, and with different cathode solution strengths, and of KC96 ozonesondes: Results of the BESOS balloon flight, submitted to *Journal of Geophysical Research*, 2007, hereinafter referred to as Deshler *et al.*, submitted manuscript, 2007) have indicated that the use of 1.0% KI cathode sensing solution can lead to an overestimation of ozone of up to 5% below 20 km, and up to 10% above 20 km. These conclusions are consistent with Boyd *et al.* [1998] who earlier found that the use of 1.0% solution in ES sondes could cause up to a 5% overestimation in midlatitude total column ozone. In addition, the field comparison (Deshler *et al.*, submitted manuscript, 2007) of ozonesondes to an ozone photometer suggested that the use of a pump efficiency correction factor [Johnson *et al.*, 2002] may unnecessarily increase ozone concentrations measured at low pressures.

[12] These results led to an extensive recomparison of the McMurdo ozone measurements with coincident remote measurements by satellite (e.g., TOMS) and a ground-based Dobson spectrophotometer. For this comparison, the McMurdo ozone measurements were reanalyzed by two methods: (1) by removing the pump efficiency correction factor and (2) by using a linear pressure-dependent transfer function (Deshler *et al.*, submitted manuscript, 2007) to convert measurements using 1.0% KI sensing solution to an equivalent measurement with 0.5% KI sensing solution. Better agreement with remote measurements is found when either method of reanalysis is used, compared to the standard analysis used in the past which includes a pump correction factor, since both methods lead to an $\sim 5\%$ decrease in column ozone. The least invasive approach is to use the 1.0% KI measurements directly with no correction for the loss of pump efficiency at low pressures. This approach was applied to all of the McMurdo ozone

measurements for this analysis, except for the 1999 measurements which were completed with a 0.5% KI cathode solution and a correction for pump efficiency. The McMurdo ozonesonde observations analyzed in this way are used as the reference for comparing with the runs, UK and EC, of the SLIMCAT model.

[13] By removing the pump efficiency correction, total column ozone is decreased by $\sim 6\%$ for the ES sondes which used 1.0% KI. On individual isentropic levels, the decrease is altitude-dependent and ranges from $\sim 2\%$ at a pressure level of 100 mbar (~ 390 K) to $\sim 12\%$ at a pressure level of 10 mbar (~ 820 K). The reason a pump efficiency correction is not required for the 1.0% measurements is not well understood, but it is thought that the loss in pump efficiency may be counteracted by an increase in iodine formation due to a secondary reaction taking place in the cathode cell that involves activated oxygen [Saltzman and Gilbert, 1959]. Up to now, this secondary reaction has not been quantified in ozonesondes.

2.2. SLIMCAT 3-D Chemical Transport Model

[14] SLIMCAT is a global off-line 3-D chemical transport model (CTM). The original version used a purely isentropic vertical coordinate and extended down to about 330 K. Recently the model has been extended to the surface using a hybrid σ - θ coordinate [Chipperfield *et al.*, 2005; Chipperfield, 2006]. SLIMCAT uses prescribed horizontal winds and temperatures specified by meteorological analyses. In the purely isentropic domain vertical advection is calculated using a radiation scheme. In the past this was the Middle Atmosphere Radiation Scheme (MIDRAD) [Shine, 1987], although recent runs have used a scheme from the NCAR Community Climate Model (CCM) [Briegleb, 1992; Feng *et al.*, 2005]. Chemical tracers are advected by conservation of second-order moments [Prather, 1986]. The model includes a detailed gas phase stratospheric chemistry scheme, and accounts for heterogeneous reactions on liquid aerosols, nitric acid trihydrate (NAT) and ice [see Chipperfield, 1999, 2003]. The model includes passive ozone (i.e., ozone advected without chemical changes) which is reset equal to the fully modeled ozone on 1 June of each year.

[15] In this study, two model runs which both have a horizontal resolution of $7.5^\circ \times 7.5^\circ$ are used. SLIMCAT run UK was forced with the UKMO meteorological analyses on 18 standard UARS potential temperature (θ) levels from 330 K to 3000 K. The model was run from 1991 to 2001 and used MIDRAD for the calculation of vertical motion. The time resolution is 24 hours. This run, performed in 2001, represents the model at that time and corresponds to the “old” SLIMCAT run (run 198) used in the Arctic study of Chipperfield *et al.* [2005]. To add ozone below 330 K, ozonesonde measurements at McMurdo were used to produce a temporally dependent column of ozone below 330 K which was added to the total ozone amounts for run UK. This resulted in a temporally dependent addition of ~ 15 –35 Dobson units between days 30–224, and ~ 35 –25 Dobson units between days 225–310.

[16] SLIMCAT run EC represents the current version of the model and was forced with ERA-40 ECMWF meteorological reanalyses on 24 σ - θ levels extending from the surface to 3000 K. The ECMWF analyses are available every six hours on the original ECMWF model levels. The

run was started in 1977 and is compared to measurements from 1989 to 2003. Above a potential temperature of 350 K, run EC uses pure isentropic levels. In this region, vertical (adiabatic) motion is calculated using the CCM radiation scheme [see *Feng et al.*, 2005]. In some instances, the ERA-40 meteorological reanalyses cause stratospheric meridional circulation that is too fast, thus this model run uses diagnosed heating rates for vertical transport [*Chipperfield*, 2006]. Run EC corresponds to SLIMCAT run (run 323) as used by *Chipperfield et al.* [2005] which gives a good simulation of Arctic ozone. Although we label the model runs by the wind analyses used to force the model (UK or EC), run EC also has improved vertical resolution, vertical velocity, chemistry and denitrification in addition to the other changes mentioned here. More information is given by *Chipperfield et al.* [2005] and *Chipperfield* [2006].

[17] Above 350 K, SLIMCAT can be compared with ozonesonde measurements on a common vertical θ coordinate. Below 350 K, run EC uses hybrid levels that vary with time. Thus the model theta levels are temporally dependent, making comparisons with ozonesondes complex. This fact coupled with our interest in the stratosphere precluded comparisons below 350 K. For comparison with the SLIMCAT model, the vertical profile O_3MR measurements were binned using the model level interfaces (potential temperature) as boundaries and then averaged within those bins to get a midpoint value. SLIMCAT has 18 levels for run UK and 24 levels for run EC, with each upper interface also being the lower interface of the level above.

3. Results

3.1. Total Column Ozone

[18] The comparison of SLIMCAT run EC (1989–2003) and run UK (1992–2001) with measured total column ozone is shown in Figure 1. Before \sim Julian day 255 (13 September), both SLIMCAT runs are often within the error limits of the sonde measurements. However, after day 255, run UK overestimates total column ozone by more than 25% in many cases, except in 2001 where agreement is good throughout the measurement period. The overestimations continue through late October in the years prior to 1997. From 1997 onward, run UK tends to agree with run EC and with the October measurements, after \sim Julian day 290 (17 October).

[19] Run EC remains in agreement with the sonde measurements throughout each annual ozone measurement period and captures the variations in total column ozone quite well, including subtle shifts, and more strikingly, large variations in amplitude across the vortex boundary over very short time periods, such as between days 270–305 in 1992 and days 285–305 in 1996. Although run EC and the measurements agree well generally, run EC shows underestimations of total ozone during some of the time periods including late 1990, 1996 and 2001 as well as most of 2003.

[20] Early and midwinter ozonesonde data are available for comparison in 1994 and 2003. In 1994, run UK shows the same general trend as in the other years with generally good agreement prior to mid-September, but a large overestimation of total column ozone after about day 255. Run EC generally overestimates total column ozone prior to \sim day 205 in 1994, but agreement improves through the rest

of the year. In 2003, run EC and the measurements show a persistent difference in total column ozone during the winter months and the ozone depletion period, although the structure of the variation is well captured. The reason for this difference is unknown. Dobson spectrophotometer measurements (Figure 1) taken by moonlight during winter 2003 agree with the ozonesonde measurements except for three Dobson measurements which are even lower than the SLIMCAT model estimates. However, the possible error on Dobson moonlight measurements is large and the error bars on the three low measurements are within the errors on the ozonesonde measurements.

[21] Overall, run EC, with improved transport and chemistry, and tropospheric ozone, provides a good simulation of column ozone variations through the austral winter/spring. Comparisons with other ozone column measurements, such as Dobson spectrophotometer measurements near McMurdo (1998 and 2003 in Figure 1), illustrate the strengths and weaknesses of such remote measurements: high time resolution after sunrise, but a lack of measurements before sunrise. In 1998, the ozonesonde and Dobson measurements match within instrumental error throughout the late winter-early spring, as a previous comparison has shown [*Kröger et al.*, 2003]. Generally, in 1998, SLIMCAT run UK underestimates column ozone during late winter (\sim days 230–255) when compared with the measurements, both in situ and remote. However, with the exception of four measurements, run UK overestimates ozone when compared with the measurements between \sim days 250–276. Agreement is good again in late October (\sim days 278–305). Run EC agrees well with the measurements throughout 1998, but with a slight underestimation in late October.

[22] As discussed in the introduction, a comparison of SLIMCAT estimates with Dobson spectrophotometer measurements at Halley Bay, Antarctica ($76^\circ S$, $333^\circ E$) indicated that SLIMCAT (also forced by UKMO winds) overestimated total column ozone throughout the study period [*Chipperfield*, 1999]. This agrees with the relationship seen here between run UK and ozone column measurements above McMurdo during the ozone hole period, although the overestimation by run UK in this study is less than in the Halley Bay study both in magnitude and duration.

3.2. Ozone Mixing Ratio (O_3MR)

[23] Total column ozone comparisons provide a picture of the overall ability of SLIMCAT to capture trends and interannual variability in the annual cycles of polar stratospheric ozone. Comparisons of modeled and measured ozone profiles provide more detail and may indicate regions in the vertical where measured and modeled ozone agree well and where there are differences. Here, the measured O_3MR from all years and all ozonesonde flights are used for comparison with the model at all possible model levels where they overlap.

3.2.1. SLIMCAT Run UK

[24] To quantitatively assess the agreement between the model and measurements, the ozonesonde measurements were binned at the vertical resolution of run UK. For all comparison points, run UK produces an average O_3MR of 2.25 with standard deviation of 1.76, while the measurements have an average O_3MR of 2.05 and standard deviation of 1.05. These values are indicative of the density of

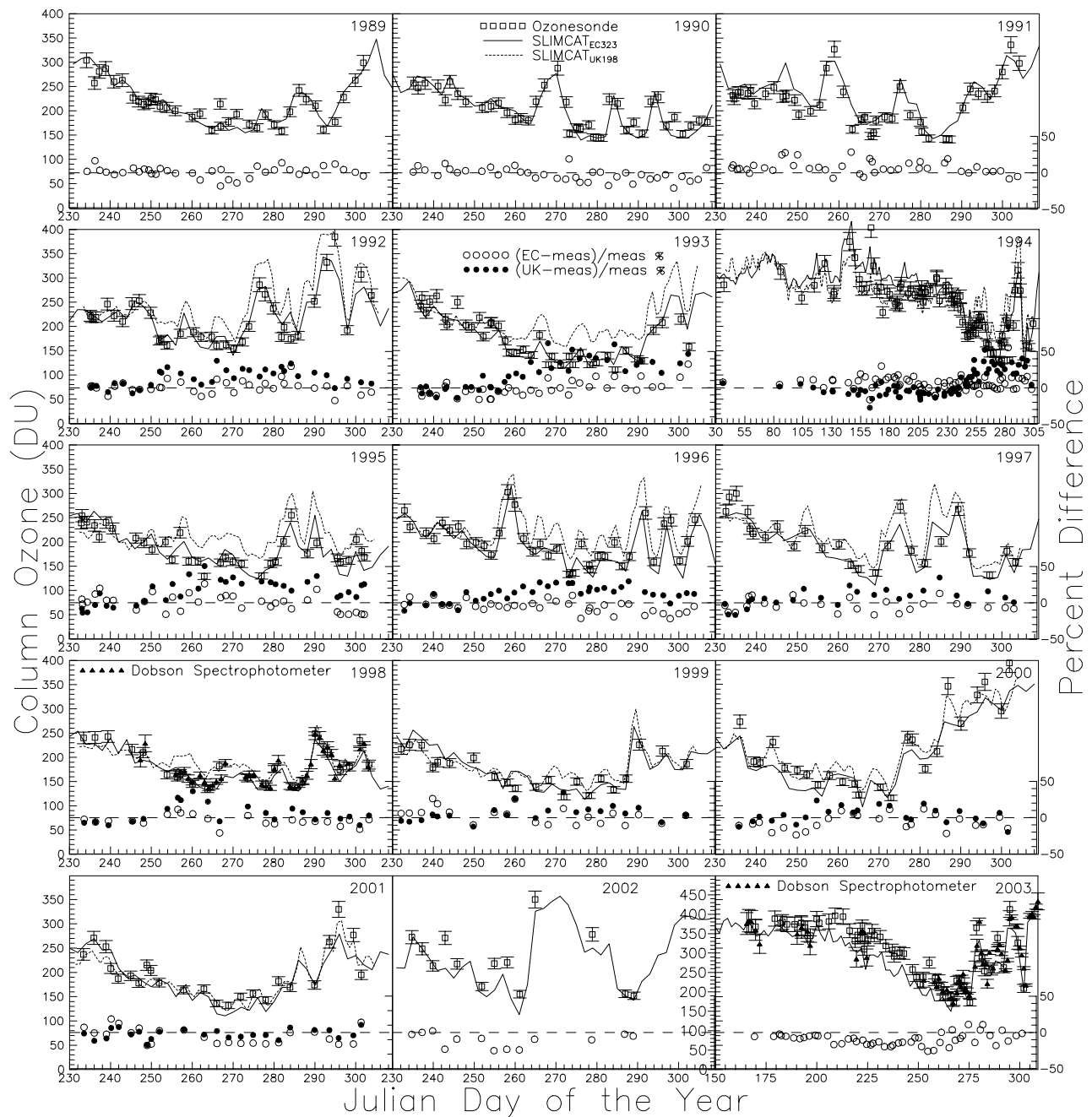


Figure 1. Total column ozone in Dobson units measured by balloon-borne ozonesondes (squares with $\pm 5\%$ error bars), and estimated with SLIMCAT runs UK (dashed line) and EC (solid line) versus Julian day for 1989–2003 above McMurdo Station, Antarctica (77.8°S , 166.7°E). Time ranges from day 230 (~ 19 August) to day 310 (~ 7 November), except in 1994 where the record starts at day 30 (31 January) and in 2003 where the record starts at day 150 (31 May). Column ozone corresponds to the left-hand y axes and ranges from 0 to 400 DU, except in 2002 where a separate axis label indicates a range of 0–500 DU. Percent differences shown at the bottom, right-hand y axes, are given by $(\text{model-measurement})/\text{measurement}$ for SLIMCAT runs UK (solid circles) and EC (open circles). Also included for 1998 and 2003 are total column ozone Dobson spectrophotometer measurements (triangles with error bars). Error bars on Dobson spectrophotometer measurements range from 1.5 to 7.5%, depending on the measurement conditions (based on *Basher* [1982]).

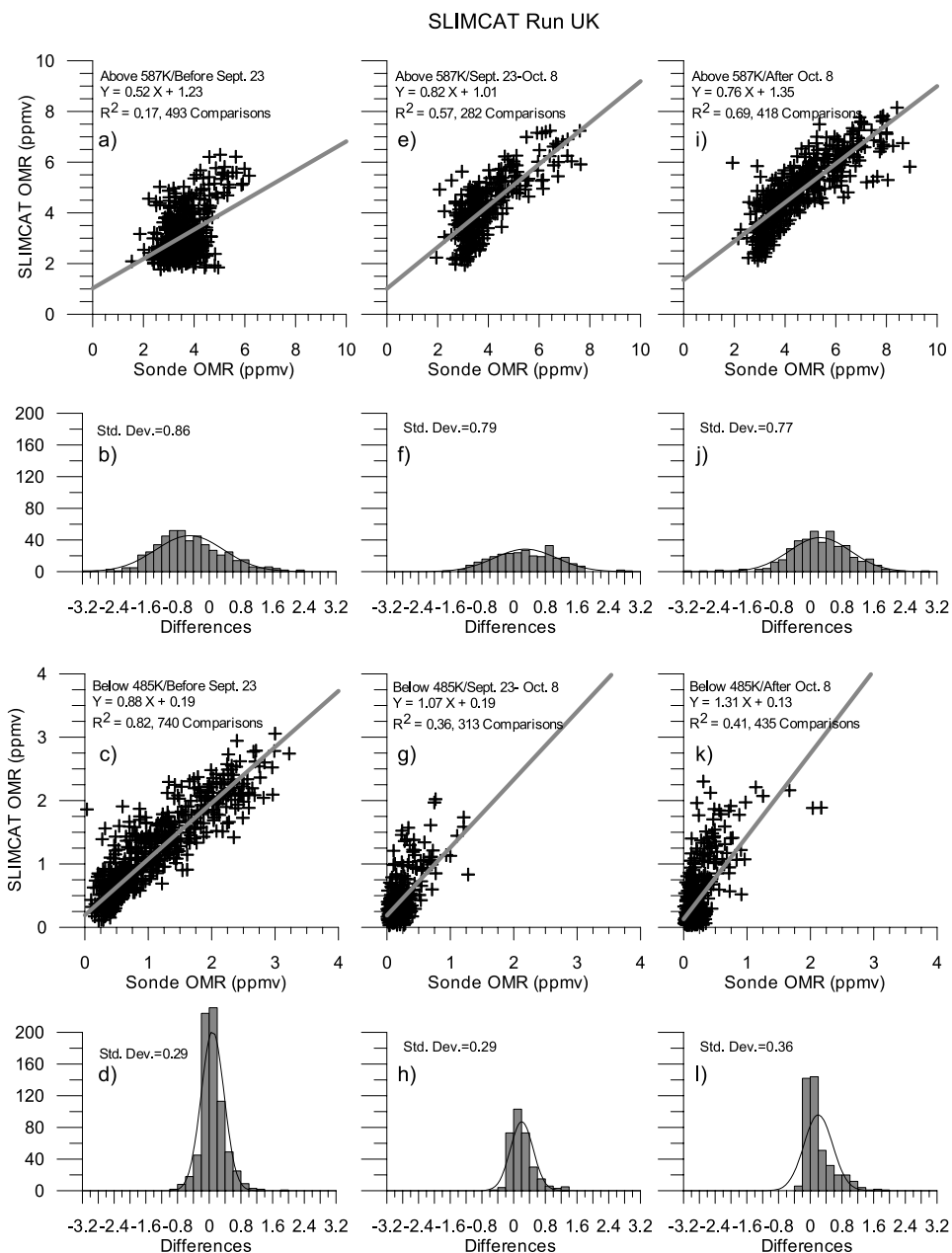


Figure 2. Statistical comparison of sonde measurements of ozone mixing ratios (O_3MR) with SLIMCAT run UK. Data are divided into subsets for comparisons (a–d) before 23 September, (e–h) between 23 September and 8 October, and (i–l) after 8 October of each year and comparisons where potential temperatures are between 330–485 K (Figures 2c, 2d, 2g, 2h, 2k, and 2l) and 587–1150 K (Figures 2a, 2b, 2e, 2f, 2i, and 2j).

measurements at low O_3MR . Linear regression of the two quantities gives: $SLIMCAT_{UK} O_3MR = 0.90(Sonde O_3MR) + 0.41$, with a correlation coefficient of $R^2 = 0.84$.

[25] To more accurately assess where/when the model and measurements agree/disagree, the data are split into subcategories of three time periods; the period when ozone loss is most rapid (before 23 September, day 265), the period of minimum ozone (23 September to 8 October, days 265–280) and after 8 October (day 280). These periods are further split into two potential temperature ranges; between ~ 330 –485 K (the region where chemical ozone depletion dominates the measurements, the chemically perturbed

region) and 587–1150 K (where chemical ozone depletion is minimal, the chemically unperturbed region), Figure 2. The isentropic layers between 485 and 587 K have been excluded because ozone in this region is controlled by a mixture of chemical depletion and transport. Linear regressions of these quantities show run UK does well in reproducing O_3MR in the chemically perturbed region prior to day 265, with high correlation and a normal distribution of differences (Figures 2c and 2d). In the chemically unperturbed region (above 587 K), the correlation is much lower (Figures 2a and 2b), but this is mostly because O_3MR varies within a narrow range. Distribution of the differences

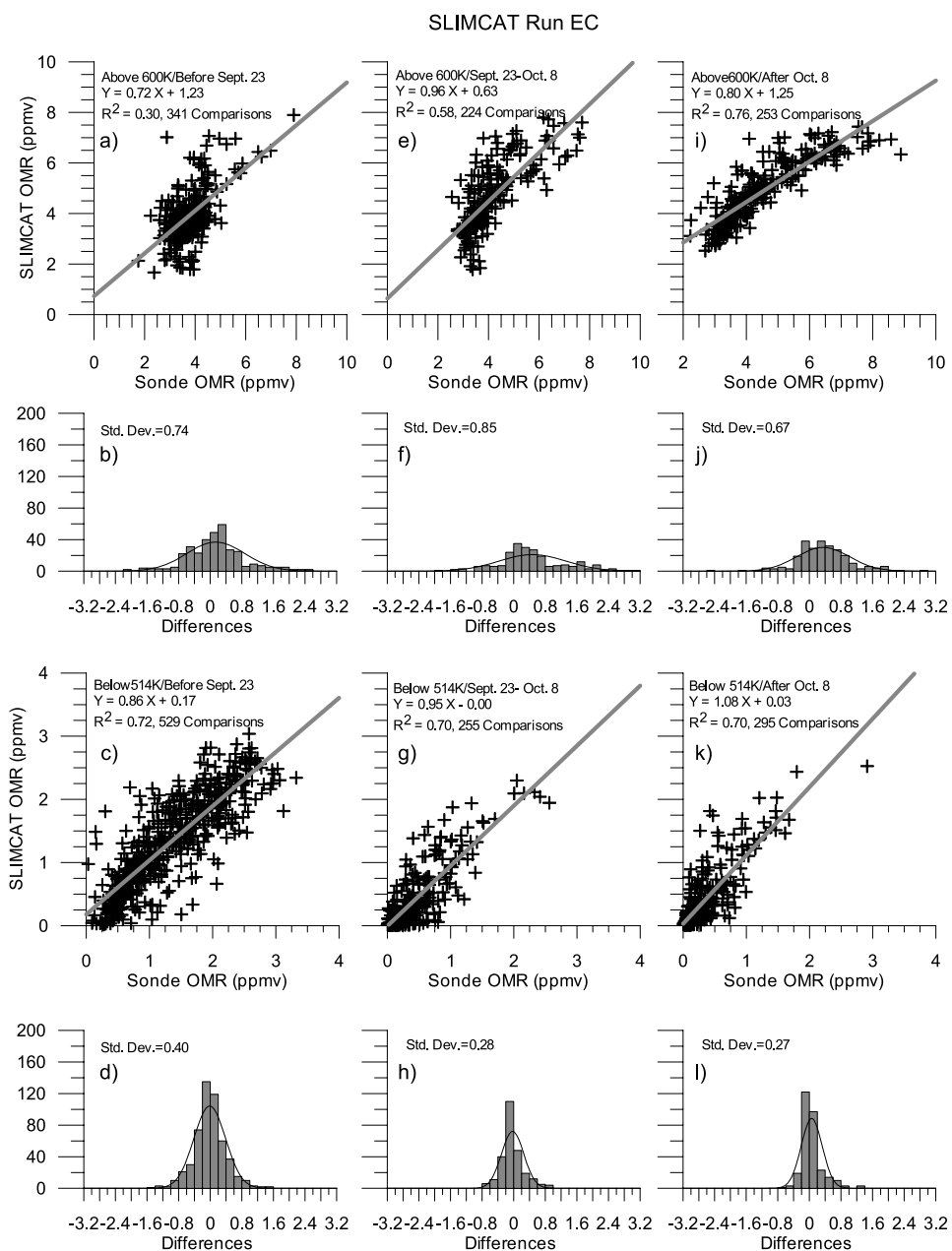


Figure 3. Same as Figure 2 for comparisons with SLIMCAT run EC, but potential temperatures range between 350–514 K and 600–1195 K.

in this case shows SLIMCAT underestimating O_3MR (Figure 2b). After day 265, the correlation improves in the upper region (Figures 2e, 2f, 2i, and 2j), but the model overestimates O_3MR in the lower region (Figures 2g, 2h, 2k, and 2l), where ozone has a low variability.

3.2.2. SLIMCAT Run EC

[26] A linear regression of O_3MR from SLIMCAT run EC and ozonesonde measurements gives: $SLIMCAT_{EC} O_3MR = 1.02(Sonde O_3MR) + 0.15$, $R^2 = 0.89$. These model estimates are very close to measured values. Run EC gives an average O_3MR of 2.44 with standard deviation of 1.81, while the measurements have an average O_3MR of 2.24 and standard deviation of 1.67. Note that these calculations for the measurements are done after they are binned according to model interface levels, thus the difference between

average and standard deviation for the measurements compared to run EC or run UK.

[27] Results for run EC were also split into the same three time periods (before 23 September, 23 September to 8 October, after 8 October), and potential temperatures between ~ 350 –514 K and ~ 600 –1195 K, Figure 3. Isentropic layers between 514 and 600 K have been excluded because of the mix of chemistry and transport controlling ozone amounts. Good correlation is seen in the chemically perturbed region (below ~ 514 K) throughout the three time periods (Figures 3c, 3d, 3g, 3h, 3k, and 3l), as well as in the chemically unperturbed region (above ~ 600 K) after 23 September when depletion is near completion (Figures 3e, 3f, 3i, and 3j), although the distribution of differences is skewed to the positive in these latter cases. Before

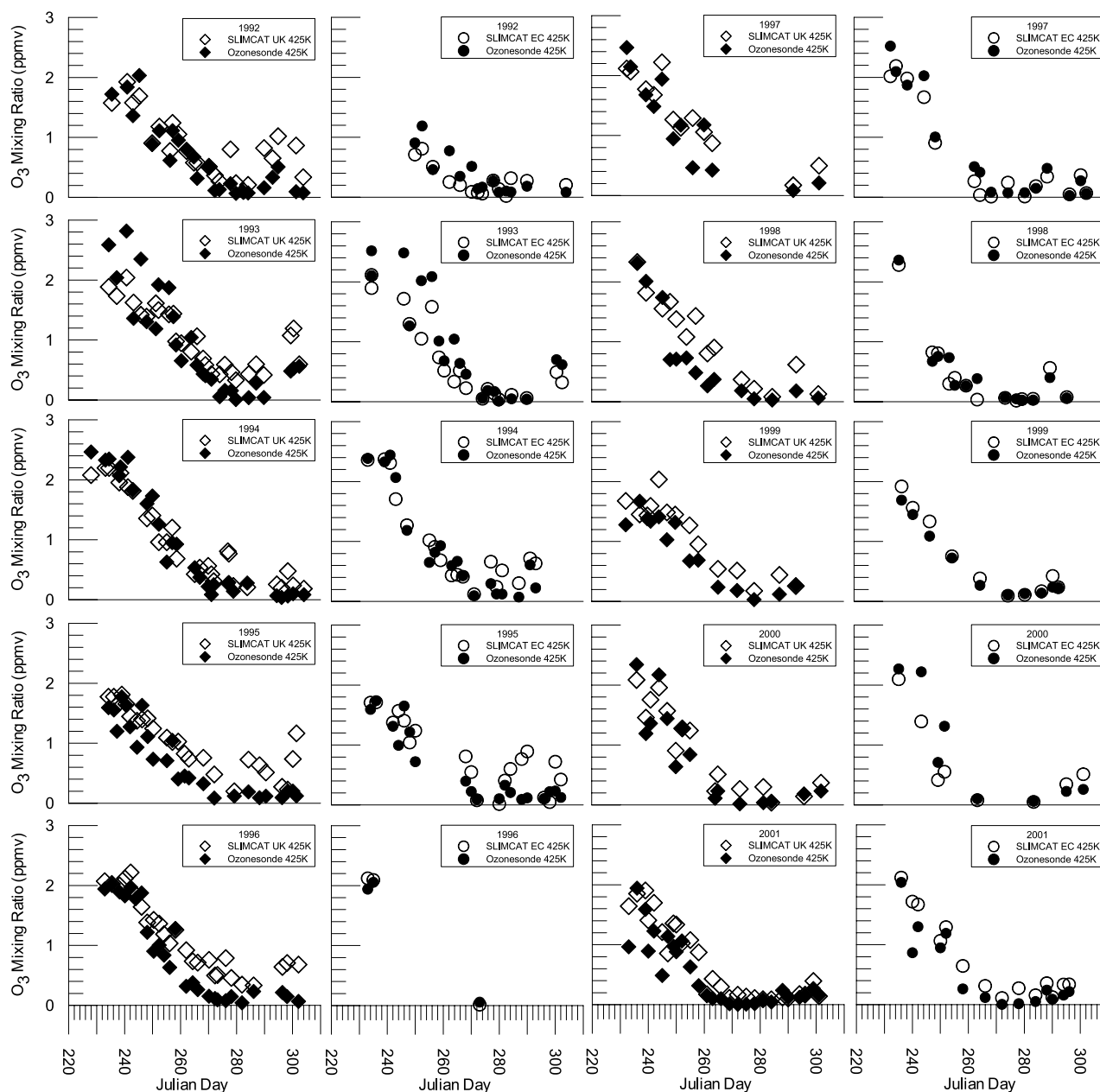


Figure 4. Ozone mixing ratios at ~ 425 K. Note the vertical resolution is half that of Figure 5.

23 September in the chemically perturbed region, the R^2 value is lower (0.72) than for run UK (0.82). While there is little change in the regression equations between the two model runs for this period, the variance is larger for run EC (Figure 3d) as indicated by the distribution of differences and standard deviations. In the unperturbed region before 23 September, although the narrow range of O_3MR again limits the correlation (Figure 3a), the distribution of the differences (Figure 3b), indicates improved agreement over the comparison with run UK (Figures 2a and 2b).

3.2.3. O_3MR at Specific Potential Temperatures

[28] Although the previous O_3MR comparisons give an overall impression of model performance, they mask all temporal information in the vertical profile. To explore this aspect of the comparisons, O_3MR at two potential temperature levels, one within the chemically perturbed region

(425 K) and one above it (690 K for run UK, 673 K for run EC), are analyzed in detail between 1992 and 2001 (Figures 4 and 5), years when both model runs are available. The limited number of matches in 1996 for run EC results from a poor temporal correspondence of sampling and model results for that year. The higher frequency of comparisons with run UK result from the higher temporal resolution of the run UK output (every 24 hours). Run EC was run with output for every other day. It is important to note that Figures 4 and 5 have different vertical resolutions to accommodate the range of ozone mixing ratios at the two different isentropic levels.

[29] At 425 K, run UK overestimates O_3MR during the depletion and ozone minimum periods in all years except 1994, 1997 and 2001 where agreement is good (Figure 4). Run EC does not show the same overestimation, except

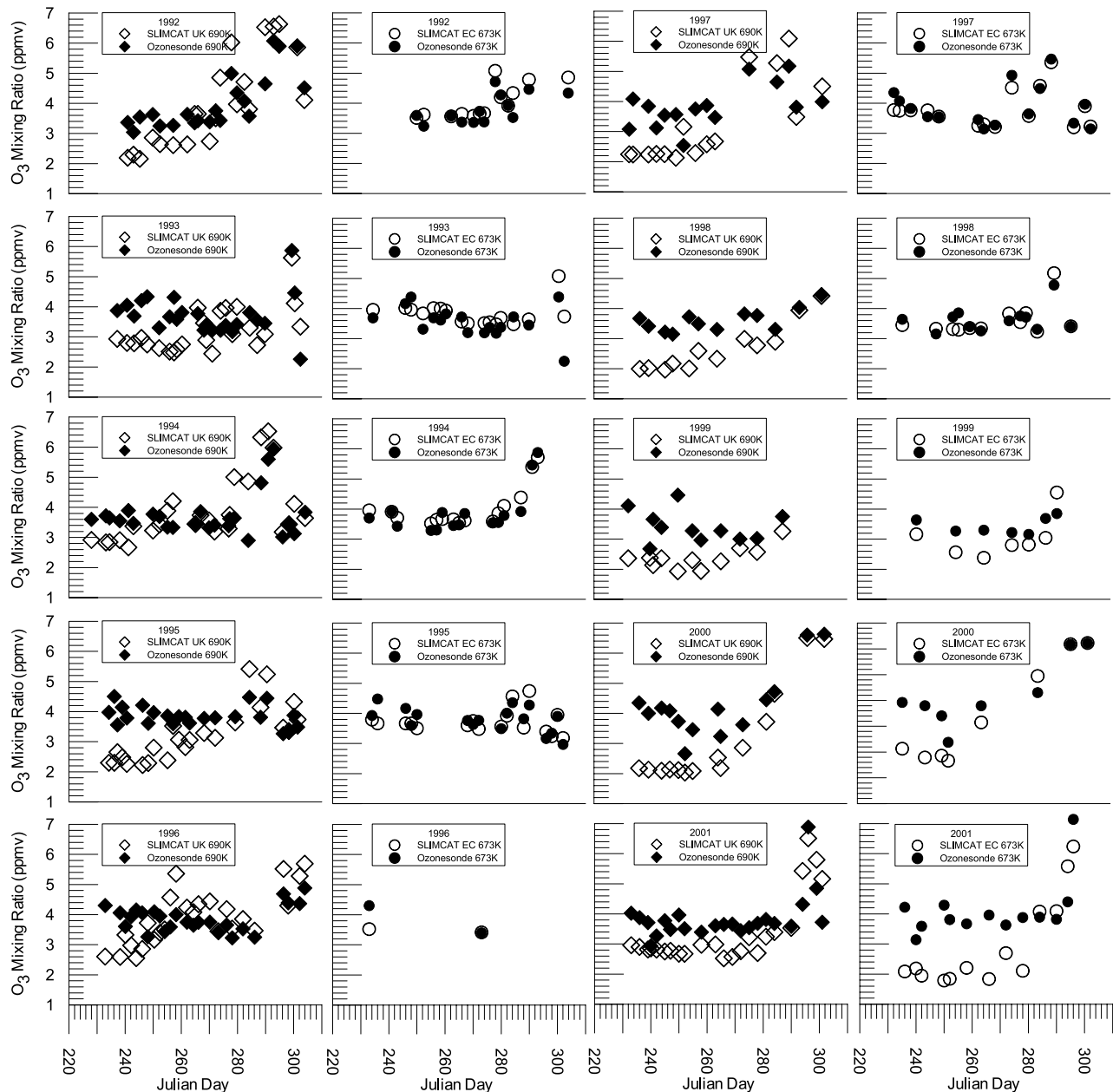


Figure 5. Ozone mixing ratios at ~ 690 K (673 K for run EC). Note the vertical resolution is twice that of Figure 4.

during late 1994 and 1995 where the model overestimates ozone in the long ozone minimum period. At 690 K, where O₃MR are less affected by chemical perturbation, run UK underestimates O₃MR through most of the comparison period (Figure 5). Exceptions to this pattern are in 1994 where run UK agrees reasonably well during the ozone minimum period, and in 1996 where it overestimates O₃MR during the same period. Agreement somewhat improves in most years late in the measurement period.

[30] Run EC shows considerably better agreement with measurements at both levels, 425 K and 673 K. Exceptions occur in 1993 at 425 K during the depletion period and in 1995 where the model shows recovery beginning prematurely (Figure 4). Another exception occurs at 673 K beginning in

1999 where the model underestimates O₃MR during the ozone depletion period (Figure 5).

4. Discussion

[31] The overestimation of total column ozone by run UK during the ozone depletion periods is consistent with previous studies, such as in Halley Bay, Antarctica [Chipperfield, 1999]. On the basis of studies in the Arctic [Feng *et al.*, 2005], these differences are thought to be due to inadequate vertical ozone transport due to an inadequate radiation formulation. The UK version is driven horizontally by UKMO analyses and vertically by the MIDRAD radiation scheme. The lowest correlation in the comparison

with Antarctic measurements shown here is in the early season above potential temperature levels of 587 K (Figures 2, 3, and 5), suggesting that the dominant problem is transport and not model chemistry.

[32] The new version of SLIMCAT, run EC, shows significantly better agreement with the Antarctic measurements than run UK. This is thought to be mainly due to changes in the dynamic and radiation schemes used in the SLIMCAT model. Horizontal transport in the EC version of SLIMCAT is driven by ERA-40 ECMWF (reanalysis) and vertical transport is driven by a GCM radiation scheme [Chipperfield *et al.*, 2005]. The finding that run EC provides a better comparison with column ozone measurements is in agreement with other published studies [Hoppel *et al.*, 2005; Chipperfield *et al.*, 2005; Singleton *et al.*, 2005; Feng *et al.*, 2005]. The higher temporal resolution of ECMWF, as well as the higher spatial resolution, contributes more accurately aged air parcels for use in the model.

[33] The agreement of SLIMCAT run EC with measured total ozone is quite reasonable. There are random differences but there appears to be no systematic divergence over the period of measurements, generally late August to late October, Figure 1. The average difference over each yearly measurement period is within $\pm 5\%$ of zero for run EC, which is comparable to the uncertainty of the measurement, except for 2002 and 2003 when the difference is approximately -10% . In contrast run UK generally overestimates the measurements after day 250, leading to average differences over the yearly measurement periods on the order of $5\text{--}15\%$. This divergence is particularly noticeable prior to 1997, after which the average difference decreases to about 5% , comparable to differences with run EC.

[34] When model estimates are broken into three time periods representing the period of rapid ozone loss, ozone minimum, and postozone minimum and potential temperature levels in the chemically perturbed region ($\sim 330\text{--}550$ K) and above it ($\sim 600\text{--}1200$ K), it is clear that both model runs do well in reproducing ozone mixing ratios in the chemically perturbed region during the period of rapid ozone loss. This good agreement is maintained for all time periods at this level in run EC, whereas run UK shows somewhat worse correlation after the period of rapid ozone loss. In the chemically unperturbed region after 23 September, both model runs display similar and reasonably good agreement with measurements; however, both model runs tend to underestimate O_3MR in the chemically unperturbed region before 23 September, although it is less pronounced in run EC. The improvement at higher isentropic levels is due to the change in radiation scheme where it has been shown previously that using a GCM radiation scheme, such as that used here, gives a much better representation of vertical transport [Feng *et al.*, 2005].

[35] While we cannot explain or investigate each observed model-measurement difference, the possibilities of temperature oscillations and inadequate chlorine activation are considered. Several studies have shown that there is a tendency for differences between measured and modeled (ECMWF or UKMO) temperatures to oscillate with altitude in the polar regions [Gobiet *et al.*, 2005; Manney *et al.*, 2003; Austin *et al.*, 2003]. To determine if such an oscillation may affect the results here, a comparison between the ozonesonde measured temperatures and meteorological

analyses temperatures (UKMO and ECMWF) was completed (not shown). While a composite of all profiles shows differences between measured and modeled temperatures which oscillate with altitude over McMurdo, there is no correlation with a similar composite of the differences in modeled and measured O_3MR . Further, during late winter/early spring, the Antarctic vortex remains continuously cold enough for PSC formation, thus making temperature oscillations less important [Hoppel *et al.*, 2005] when considering chlorine activation.

[36] Another consideration is accuracy of the activation of chlorine within the model since some models have proven to have insufficient chlorine loading [e.g., Lemmen *et al.*, 2006]. This does not seem to be the case for SLIMCAT. Preliminary comparisons between run EC and satellite measurements made by the microwave limb sounder (MLS) and Atmospheric Chemistry Experiment (ACE) from two Arctic and two Antarctic winters show that the SLIMCAT model overestimates chlorine activation (M. L. Santee *et al.*, A study of stratospheric chlorine partitioning based on new satellite measurements and modeling, submitted to *Journal of Geophysical Research*, 2007). Both the spatial extent and the magnitude of reactive chlorine were overestimated, largely, it is thought, because of a simplified PSC parameterization in the model. If overestimated active chlorine in the model was the main cause of observed differences in ozone mixing ratios, the result would be underestimations of measured ozone, particularly between days 240 and 280. In general, this is not observed for run EC, while run UK consistently overestimates ozone in the chemically perturbed region (Figures 1 and 4). Since the model is not underestimating ozone, it is likely that subsidence in the model is too strong and thus masking the effect of chlorine overactivation. This is also addressed by Santee *et al.* where MLS N_2O measurements are used to show that the model calculates diabatic descent that is too strong.

5. Conclusions

[37] This study has presented a detailed comparison between total column ozone and ozone mixing ratios modeled by the SLIMCAT CTM with measurements over McMurdo Station, Antarctica, for 1989–2003. The measurements were compared with two model runs; a run using the current version of the model (run EC) which has been shown to perform well in the Arctic, and an older version (run UK) typical of the version that has been used in several previous studies [e.g., Chipperfield, 1999; Sinnhuber *et al.*, 2000; Kilbane-Dawe *et al.*, 2001].

[38] Run EC provides good estimates of total column ozone during the annual period of high ozone depletion (after \sim day 265), but occasionally deviates from the measurements in late winter during the onset of ozone depletion (Figure 1). It also shows good agreement with measured ozone mixing ratios, especially in the lower stratosphere where chemical depletion is the main controller of ozone concentration. Run UK tends to overestimate total column ozone (by up to 20%) during the austral ozone depletion period, and shows less agreement with measured ozone mixing ratios. The differences between run UK and run EC are believed to arise in large part because of differences in the meteorological analyses (UKMO and ECMWF) and

radiation schemes used (MIDRAD and GCM). Run EC has improved vertical velocity due to the use of the GCM radiation scheme and improved vertical and temporal resolution due to the use of the ERA-40 ECMWF (re)analyses. Similar conclusions were reached by Hoppel *et al.* [2005].

[39] The current version of the model (run EC) reproduces well the overall seasonal cycle of O₃, including the annual period of depletion. It can effectively estimate total column ozone minima, and ozone fluctuations within the polar vortex when annual ozone depletion is greatest, as well as ozone mixing ratios within the chemically perturbed region of ~12–20 km. This is encouraging on the basis of a multidecadal simulation where all the model chemical tracers depend on the long-term circulation and the chemical depletion depends on appropriate parameterizations within the model. This supports previous studies in the Arctic which show that the current model parameters capture the transport and chemistry of Arctic ozone depletion [Feng *et al.*, 2005]. These findings support the use of chemical tracer models as benchmarks for predictions made by chemistry-climate models [e.g., Austin *et al.*, 2003; Eyring *et al.*, 2006]. To understand differences which arise for specific days and years requires additional sensitivity tests of various model parameters and meteorological analyses, and warrant further investigation.

[40] **Acknowledgments.** This research was supported by the National Science Foundation under grant OPP0230424. Appreciation goes to the University of Wyoming Antarctic field teams 1989–2003 and to the NZ Scott Base science technicians for operation of the Dobson spectrophotometer. Work at the Jet Propulsion Laboratory, California Institute of Technology, was done under contract with NASA.

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