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[1] This paper investigates the impact of circulation changes in a changed climate on the exchange of ozone between the stratosphere and the troposphere. We have identified an increase in the net transport of ozone into the troposphere in the future climate of 37%, although a decreased ozone lifetime means that the overall tropospheric burden decreases. There are regions in the midlatitudes to high latitudes where surface ozone is predicted to increase in the spring. However, these increases are not significant. Significant ozone increases are predicted in regions of the upper troposphere. The general increase in the stratospheric contribution (O_3s tracer) to tropospheric ozone in the climate changed scenario indicates that the stratosphere will play an even more significant role in the future. *INDEX TERMS:* 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0325 Atmospheric Composition and Structure: Evolution of the atmosphere; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS:* stratosphere-troposphere exchange, tropospheric ozone, climate change, tropospheric chemistry, chemistry transport modeling

1. Introduction

[2] Historically stratosphere-troposphere exchange was considered the most important source of ozone throughout the troposphere. It is now known that anthropogenic activities over the last hundred years or so have changed the chemical composition of the troposphere. The tropospheric ozone burden has increased due to enhanced emissions of precursor species such as nitrogen oxides (NO_X), methane, carbon monoxide and other volatile organic compounds. *Volz and Kley* [1988] and *Marenco et al.* [1994] have shown that lower tropospheric ozone concentrations have more than doubled since preindustrial times, although there are questions about the validity of the earliest ozone measurements. *Stevenson et al.* [1998a] estimate that the radiative forcing due to increases in tropospheric ozone between 1860 and 1990 was 0.29 Wm^{-2}. Model predictions of future tropospheric ozone changes and radiative forcing have initially focused on projections of precursors emissions [*Stevenson et al.*, 1998a; *Intergovernmental Panel on Climate Change* (*IPCC*), 1996]. More recently *Johnson et al.* [1999], *Grewe et al.* [1999], and *Stevenson et al.* [2000] have used three-dimensional models to show that climate changes reduce ozone concentrations. This reduction is primarily through an increase in specific humidity leading to a greater flux through the reaction of $O(^1D)$ with water vapor.

[3] Input from the stratosphere is a significant source of ozone in the troposphere. *Collins et al.* [2000] give estimates from a range of models of 340–930 Tg yr^{-1} compared to a photochemical production of 2820–4190 Tg yr^{-1}. This source is obviously most significant in the upper troposphere, and this region is where ozone is most effective as a greenhouse gas. Predictions of future radiative forcing from ozone depend on accurate simulations of the upper tropospheric ozone concentrations, and hence on accurate simulations of the cross tropopause transport of ozone. Not only is ozone a greenhouse gas, but at the surface it is injurious to health [*World Health Organization* (*WHO*), 1987]. Under certain conditions ozone can be transported from the stratosphere down to the lower troposphere [*Zanis et al.*, 2003]. Predictions of future surface ozone require an understanding of the effects of climate change on this transport. *Roelofs and Lelieveld* [1997] assessed the contribution of the stratosphere to tropospheric ozone using a global chemistry transport model. They concluded that on average 40% of the tropospheric ozone originated in the stratosphere. At the surface the stratospheric contribution was between 10 and 60%. The study presented in this paper will address the change in the stratospheric contribution to tropospheric ozone due to changes in circulation in a future climate scenario.

[4] The bulk transport of air downward through the extratropical tropopause is determined by the Brewer-Dobson circulation in the downward control principle [*Haynes et al.*, 1991]. This circulation is driven by Rossby wave

action in the extratropical middle atmosphere. *Butchart and Scaife* [2001] find an increase in the mass exchange between the stratosphere and troposphere in a changed climate. On a local scale, cross-tropopause transport of ozone is determined by synoptic-scale disturbances in the tropopause regions [*Stohl et al.*, 2003]. Transport of ozone from the upper troposphere down to the lower troposphere can occur at smaller scales along fronts or in convective systems.

2. Model Description

2.1. Transport Scheme

[5] The model used in this study (STOCHEM) is a three dimensional global chemistry transport model with a Lagrangian advection scheme. It has previously been described by *Collins et al.* [1997], *Stevenson et al.* [1998b] and *Collins et al.* [2002]. The model simulations of ozone and carbon monoxide have been compared against others given by *Kanakidou et al.* [1999a, 1999b]. The cross tropopause transport has been compared against other models by *Meloen et al.* [2003]. STOCHEM fits well within the range of the modeled cross tropopause fluxes. A detailed comparison with model and measurement data for a stratospheric intrusion is given by G.-J. Roelofs et al. (Intercomparison of tropospheric chemistry models: Simulated ozone transports in a tropopause folding event, submitted to Journal of Geophysical Research, 2003). This shows that the STOCHEM model performed well in simulating the observed ozone evolution. Details of the STOCHEM model used in this study are unchanged from those given by *Collins et al.* [2002] except where described below. In this model 100 000 air parcels are advected around the globe between the surface and 10 hPa, each carrying the mixing ratios of 70 chemical species. The chemistry transport model is integrated into the Met Office climate general circulation model (GCM) called the Unified Model [*Cullen*, 1993; *Johns et al.*, 1997]. It uses the generated meteorological fields from the GCM (winds, temperatures, convective diagnostics, precipitation rates) to advect the Lagrangian parcels and act on their contents. The chemistry model is called every 3 hours by the GCM but uses a 1 hour advection time step. The Lagrangian advection is by 4th order Runge-Kutta integration, using linear interpolation to derive wind fields at intermediate times. The parcels are constrained to remain below 10 hPa. This constraint may cause the parcels to move through the upper stratosphere too quickly, but should have little effect on the cross tropopause flux as this is controlled by the downward control principle.

[6] The Unified Model (UM) is a grid point model. The version used for this study (HadAM4) is an atmosphere-only model, with a horizontal resolution of $3.75° \times 2.5°$, and 38 vertical levels. The top level is centered on 4.6 hPa. This top might not be high enough to represent the planetary wave breaking accurately. It is not obvious what effect this would have on driving the stratospheric circulation. However, the fact that we get similar results to those with a vertically extended version of the same model [*Butchart and Scaife*, 2001] gives us confidence in our results. *Rind et al.* [1999] found that in their model, stratosphere-troposphere exchange was more sensitive to the height of the model top than to the vertical resolution near the tropo

pause. However, their greatest number of levels was only 23. *Austin et al.* [1997] compared a 19 level version of the UM with a 4.6 hPa top to a 49 level version with a 0.1 hPa top. They concluded that the main effect on transport across the 100 hPa surface was due to the vertical resolution in that region, rather than the position of the model top.

[7] Currently the coupling between the GCM and the chemistry model is one-way. None of the chemical fields from the chemistry model are passed back to the GCM.

2.2. Chemistry

[8] The chemistry scheme has been described previously by *Collins et al.* [1999]. The main difference is that the model top (beneath which the parcels are constrained) has been raised from ∼100 hPa to 10 hPa and so now extends well into the stratosphere. The set of chemical reactions we use in the model has been chosen to simulate the photochemistry of the troposphere, but is not designed to simulate stratospheric chemistry. We therefore set all chemical reactions to zero above the tropopause and relax the ozone values toward a stratospheric ozone climatology [*Li and Shine*, 1995] with a 20 day relaxation time. Varying the relaxation time by a factor of two was found to have little effect on the resulting ozone profiles. The World Meteorological Organization definition of the tropopause height is used [*World Meteorological Organization (WMO)*, 1986]. A process similar to that for ozone is carried out for oxides of nitrogen (NO_Y). We derive a stratospheric NO_Y concentration by scaling the ozone climatology by 0.001 (kg (N)/ kg(O_3)) [*Murphy and Fahey*, 1994]. We assume that all the NO_Y in the stratosphere is in the form of HNO_3. This assumption does not affect the tropospheric chemistry since HNO_3 is quickly photolyzed to NO_2 in the upper troposphere and thence partitioned amongst the other NO_Y species. To trace the stratospheric ozone through the model, an additional tracer O_3s is added after the manner of *Roelofs and Lelieveld* [1997]. This is formed and removed in the stratosphere as the O_3 is relaxed toward the climatology, but has no other production routes. O_3s is destroyed in the troposphere at the same rate as O_3, except that the reactions with NO and NO_2 are excluded.

[9] The relaxation toward the stratospheric climatologies is described below.

$$\Delta O_3 = \overline{O}_3 - X_i(O_3) \qquad (1)$$

$$\Delta HNO_3 = \overline{HNO_3} - X_i(HNO_3) \qquad (2)$$

$$X_i(O_3) \rightarrow X_i(O_3) + \Delta O_3 \times \Delta t/\tau \qquad (3)$$

$$X_i(O_3s) \rightarrow X_i(O_3s) + \Delta O_3 \times \Delta t/\tau \qquad (4)$$

$$X_i(HNO_3) \rightarrow X_i(HNO_3) + \Delta HNO_3 \times \Delta t/\tau \qquad (5)$$

where $X_i(O_3)$, $X_i(O_3s)$ and $X_i(HNO_3)$ are the ozone, ozone(stratospheric) and nitric acid concentrations of parcel i, $\overline{O}_3$ and $\overline{HNO_3}$ are the climatological stratospheric ozone and nitric acid concentrations at the location of parcel i, Δt
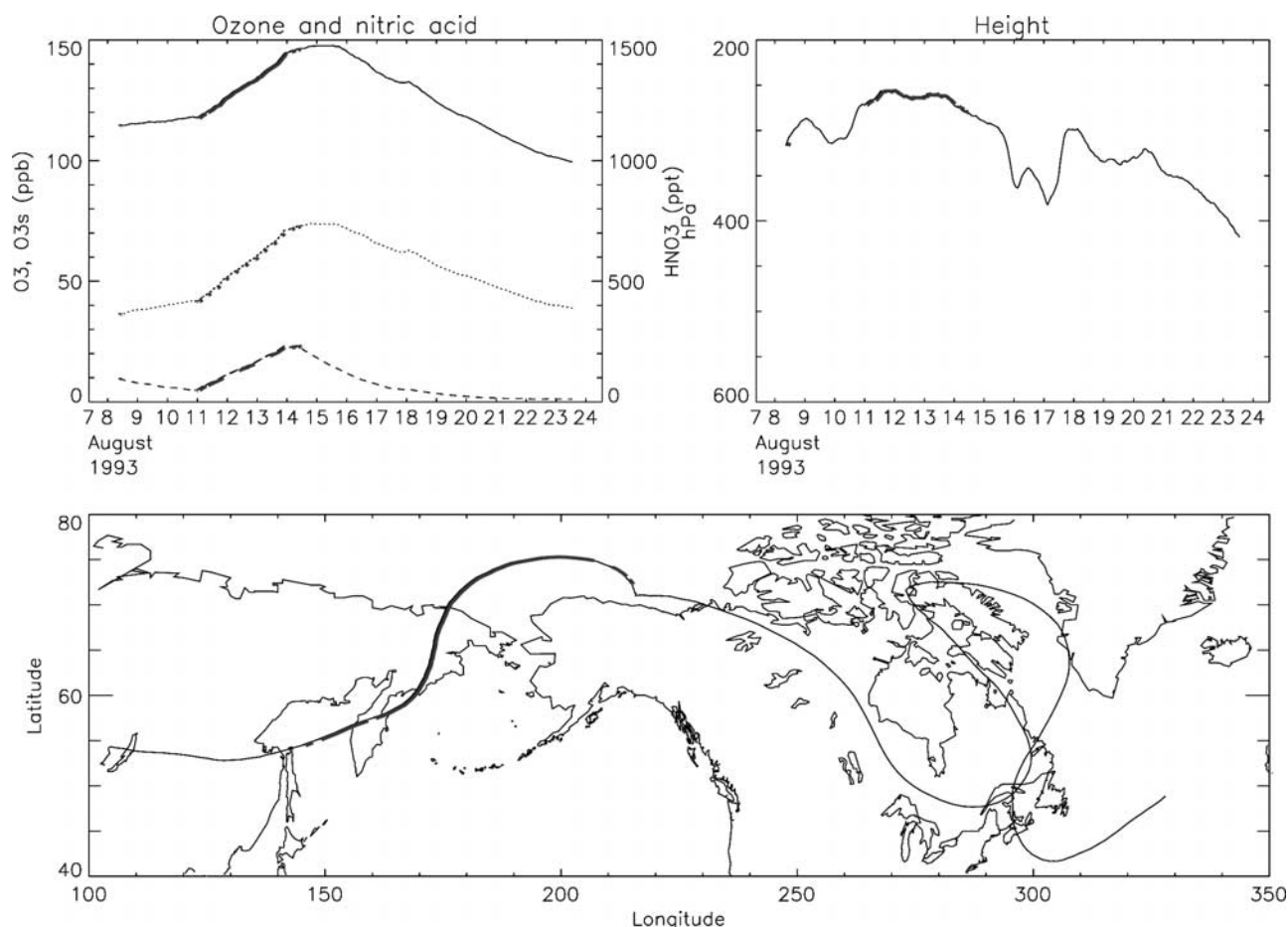
Figure 1. An example of a parcel trajectory showing its O₃ (solid), O₃s (dotted) and HNO₃ (dashed) concentrations, and height. The thicker line segments indicate when the air parcel was above the tropopause.

is the advection time step and τ is the relaxation time (20 days). Figure 1 shows an example of the changes to O_3, O_3s and HNO_3 on an air parcel as it travels up into the lower stratosphere and back to the upper troposphere. The parcel picks up 30 ppb of O_3 and O_3s, and 180 ppt of HNO_3 over 80 hours and brings these elevated concentrations down into the troposphere. As the parcel descends further, these concentrations decrease due to photochemical destruction leading to an irreversible transport of O_3 and HNO_3 from the stratosphere to the troposphere. The relatively high parcel O_3 concentration before reaching the stratosphere is due to the parcel receiving a pulse of NO_X several days previously, either from lightning or aircraft. The non-zero O_3s concentration is from recent previous incursions into the stratosphere.

3. Model Results

[10] The model was run twice with six months spin-up followed by a further four years integration. The dates covered were July 1990 to December 1994 and July 2090 to December 2094. Four years was chosen to give some idea of the statistical significance of the results. A longer integration would reduce the statistical uncertainty but in that situation systematic uncertainties due to the model

formulation would dominate. The driving GCM was run in an atmosphere-only mode. Monthly varying sea surface temperatures were taken from a previous 110 year coupled ocean-atmosphere run starting in 1989 with green house gas concentrations varying according to the A1FI scenario provided by SRES. This scenario "Fossil Fuel Intensive" was chosen as it has the greatest CO_2 rise of all the SRES scenarios, reaching 930 ppmv by 2100. The methane mixing ratio reaches 3.3 ppmv and N_2O reaches 0.465 ppmv. The 2100 global mean surface temperature change in the coupled run, relative to 1860, is 5K. This temperature rise generally increases from south to north and is larger over the continents than the oceans, except for the Arctic Ocean which has the largest surface temperature rise of all. *Rind et al.* [2002] show that the stratospheric circulation is more sensitive to surface temperature increases in the tropics compared to the extratropics.

[11] The meteorological starting conditions for the GCM in July 1990 and July 2090 were also taken from the coupled run. It should be emphasized that the simulations covering 1990 to 1994 do not attempt to reproduce the observed meteorology for those years, simply that they correspond to an atmosphere with CO_2 concentrations appropriate for that time period. The climatology for ozone above the tropopause is representative of the 1990s and is
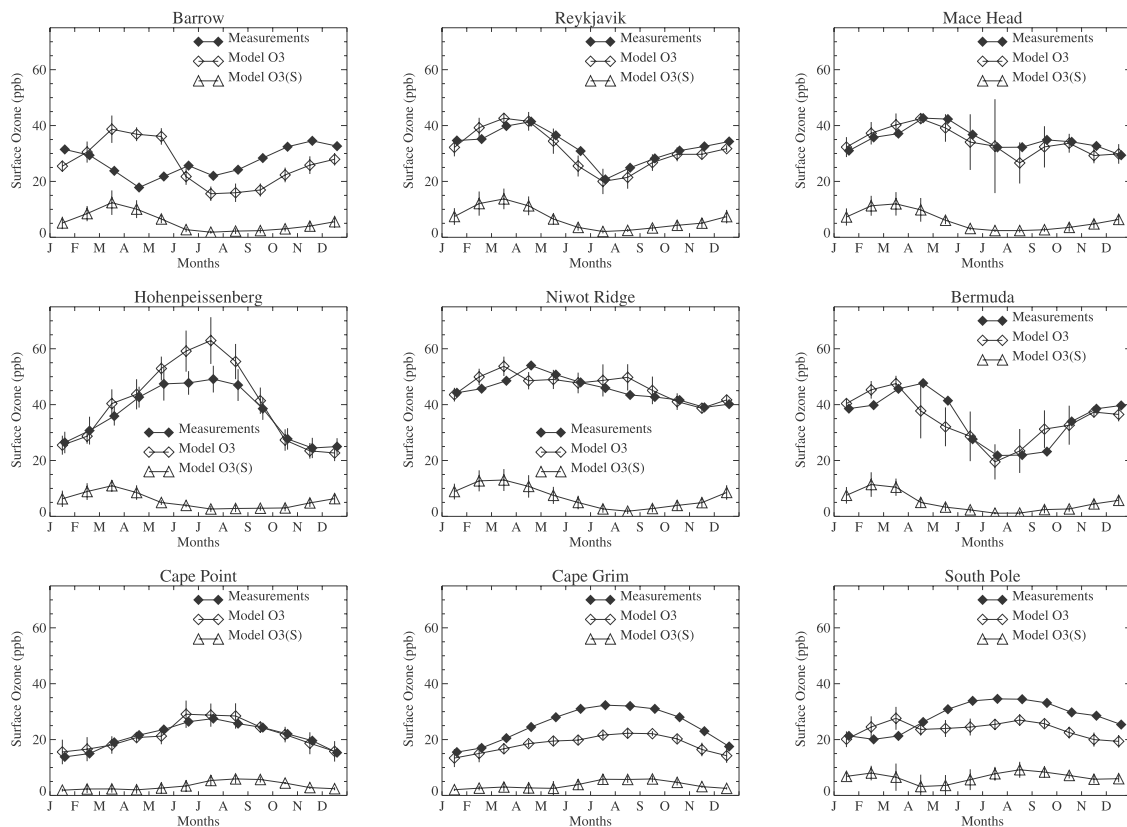
Figure 2. Simulated (open diamonds) and measured (solid diamonds) surface ozone concentrations at selected sites. Also shown are the concentrations of the modeled O₃s tracer (triangles). The model data are averages over 1991–1994. The measurement data are taken from *Logan* [1999], which include data from *Oltmans and Levy* [1994] (Barrow, Reykjavik, Mace Head, Bermuda, Cape Point, Cape Grim, South Pole), and cover varying time periods.

used for both simulations with no attempt to account for future stratospheric ozone depletion or recovery. The emissions and initial concentrations of ozone precursors are kept at 1990 levels for both runs. For a list of the emissions used, see *Collins et al.* [1999].

3.1. Comparison With Observations

[12] Figures 2 and 3 show comparisons between the simulated ozone, averaged over 1991 to 1994, and measurements from *Logan* [1999] (including data from *Oltmans and Levy* [1994]). Both O₃ and O₃s are plotted for the model. The surface results show a pretty good simulation of the seasonal cycles. The most obvious exception is at Barrow, where the model simulates a springtime maximum in ozone in contrast to the observed springtime minimum. The minimum is caused by ozone destruction by halogen chemistry in the boundary layer after the Arctic winter. No halogen chemistry is simulated in the model. There is an overestimate of the surface concentrations in the austral winter, both at Cape Grim and the South Pole, although the agreement at Cape Point is very good. The Cape Grim measurements have been filtered according to wind sector to exclude air masses originating from over the polluted continent, and so might be expected to under represent the monthly averaged values. Summertime ozone is substantially overpredicted at Hohenpeissenberg due to too much photochemical production. The emission grid used in our

model is 5°×5° which has the effect of spreading out the emissions from German cities and cannot distinguish between the industrial and rural areas.

[13] The O₃s concentrations are much lower than the full ozone concentrations as the majority of surface ozone comes from photochemical production rather than transport from the stratosphere. The summertime concentrations are only of the order of 2–3 ppb, however there are springtime maxima in O₃s at the northern hemisphere sites of 10–15 ppb. The effects of these maxima can also be nicely seen in the increases in full ozone, both in the model and in the measurements, however the modeled ozone often peaks one month earlier than the measurements. The springtime O₃s peak is due to a competition between transport from the stratosphere which peaks in the late spring to early summer and the ozone lifetime which is longest in the winter and shortest in the summer. In the early spring, the model simulates some ozone transport to the northernmost sites from locations further south where photochemical production has already started. This transport seems to be too efficient, probably due to too little surface destruction of the ozone en route, causing an overestimate of the early spring ozone. It is obvious from Figure 2 that transport from the stratosphere makes a significant contribution to the northern midlatitude spring ozone maximum, but it is not the only process making a contribution [*Monks*, 2000]. Net ozone production in the boundary layer is also a maximum in the
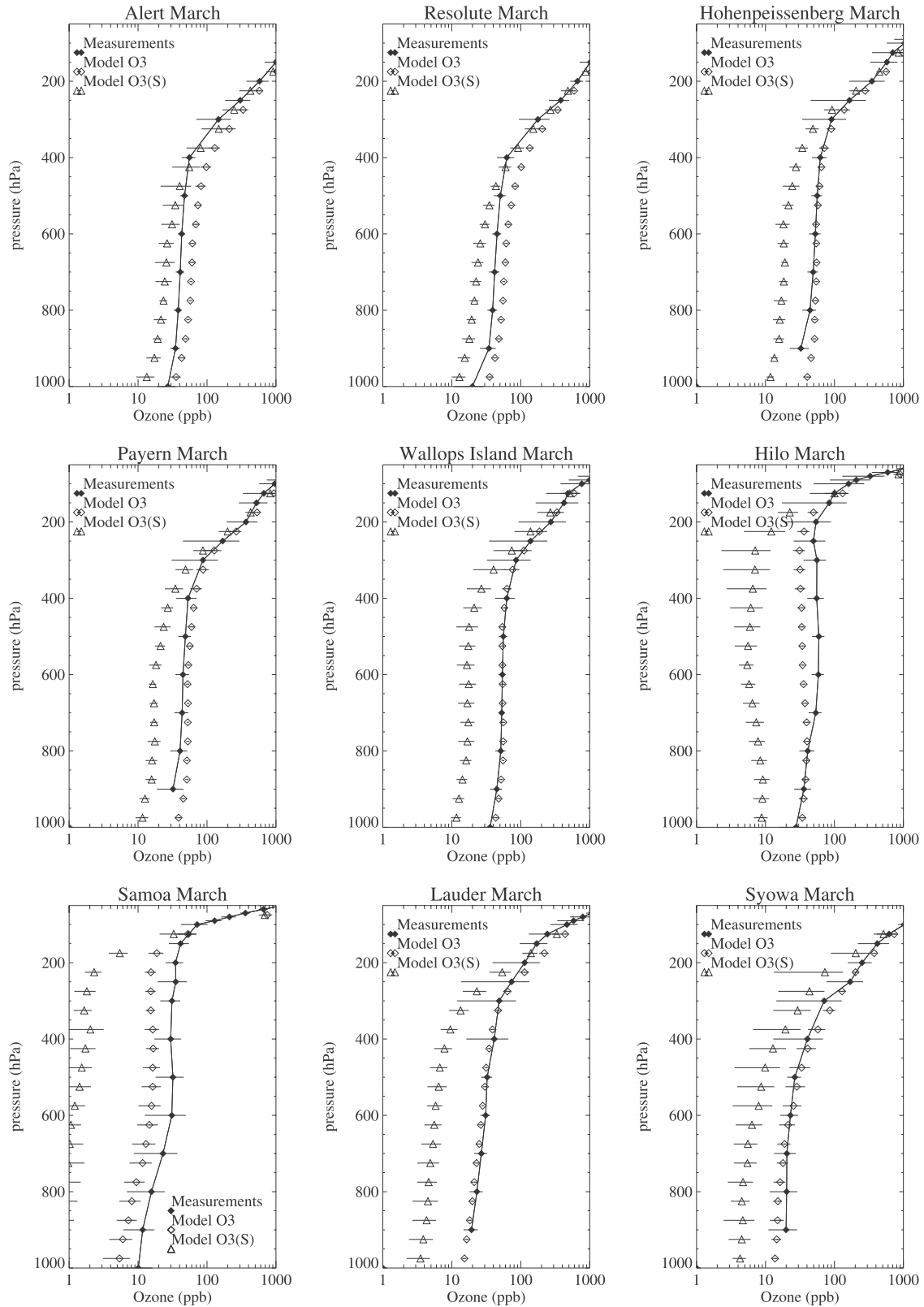
Figure 3. Simulated (open diamonds) and measured (solid diamonds) ozone profiles at selected sites for the month of March. Also shown are the concentrations of the modeled O_3s tracer (triangles). The model data are averages over 1991–1994. The measurement data are taken from *Logan* [1999] and cover varying time periods.
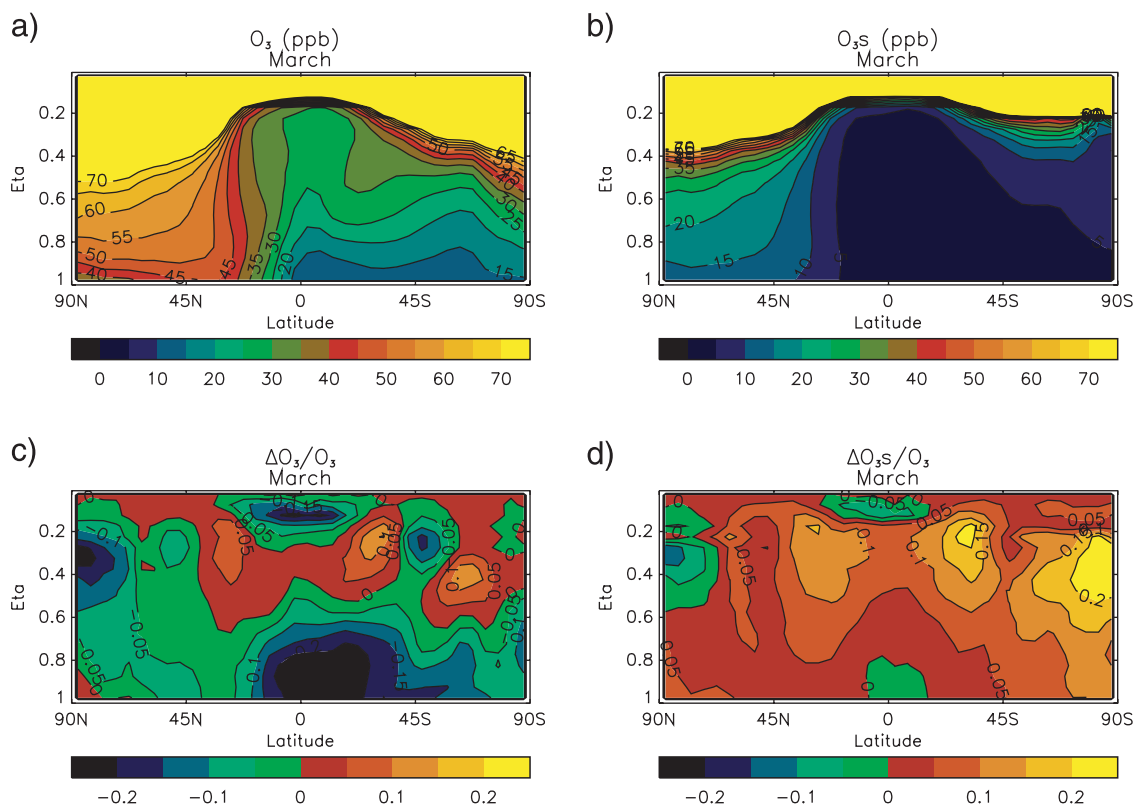
Figure 4. Model zonal mean O_3 concentrations (a) and O_3s concentrations (b), averaged over 1991– 1994. Units are ppb. Zonal mean changes between 1991–1994 and 2091–2094 in O_3 (c) and O_3s (d). Changes are expressed as a fraction of the 1991–1994 full ozone. The vertical coordinate is a hybrid pressure coordinate approximately equal to pressure/surface pressure.

spring, since at this time the main production term ($NO_2+hv{\rightarrow}NO+O(^3P)$) is increasing more rapidly with season than the main loss term ($O_3+hv{\rightarrow}O(^1D)+O_2$ followed by $O(^1D)+H_2O{\rightarrow}2OH$). This, combined with a build up of ozone precursors (hydrocarbons and oxides of nitrogen (NO_Y)) over the winter in the midlatitudes to high latitudes, leads to a photochemical spring ozone maximum. The effect of this can be seen at Mace Head and Bermuda where the modeled full ozone peaks a month later than the stratospheric ozone, although this peak is still a month earlier than the measurements.

[14] The agreement between simulated ozone and ozonesondes in March shown in Figure 3 is good (although note the logarithmic scale). The agreement above the tropopause might be expected to be exact, since the model is relaxed toward the climatology. The reason for differences above the tropopause (for example at Lauder) is that the years over which the sonde data have been averaged, and the actual stations used are different for the climatology [*Li and Shine*, 1995] and the plotted data [*Logan*, 1999]. Concentrations at Alert and Resolute seem to have been over predicted by 50–100% in the lower stratosphere/upper troposphere and 10–20% in the lower troposphere. This could again be due to discrepancies between the climatology and the ozone sondes at high latitudes or a problem modeling the chemistry in this region. Midtropospheric to upper tropospheric concentrations are underestimated in the tropics in March although other months (not shown) give better agreement. This could reflect uncertainties in the modeling of the

seasonality of the ozone precursor emissions from lightning and biomass burning.

[15] The month of March was chosen for the comparison as this is the time of greatest impact of the stratosphere on tropospheric ozone at the northern sites. At the high latitudes (Alert, Resolute) O_3s makes up around 50% of the full ozone in the midtroposphere. At the midlatitudes (Hohenpeissenberg, Payerne, Wallops Island) the proportion is around 30% and decreases in the tropics to 15% at Hilo and less than 10% at Samoa. The stratospheric influence is less at the southern hemisphere sites (Lauder and Syowa) since this is the austral autumn.

3.2. Effect of Climate Change

[16] The aim of this work is to study the effect of the changed circulation due to climate change on stratosphere-troposphere exchange of ozone. Ideally we would like to keep all inputs to the model the same in the future run except for the wind fields. Unfortunately, not only does climate change due to increasing CO_2 affect the wind fields, it also changes other meteorological factors such as temperature, humidity, clouds and precipitation. All these will affect the modeled ozone distributions. The dominant effect is the increase in humidity in the 2090s meteorology which causes a more rapid ozone loss through the $O(^1D)+H_2O{\rightarrow}2OH$ reaction. A further problem when trying to isolate the effects of circulation changes is the specification of the stratospheric ozone climatology. This climatology was deliberately kept fixed for this experiment, but obviously
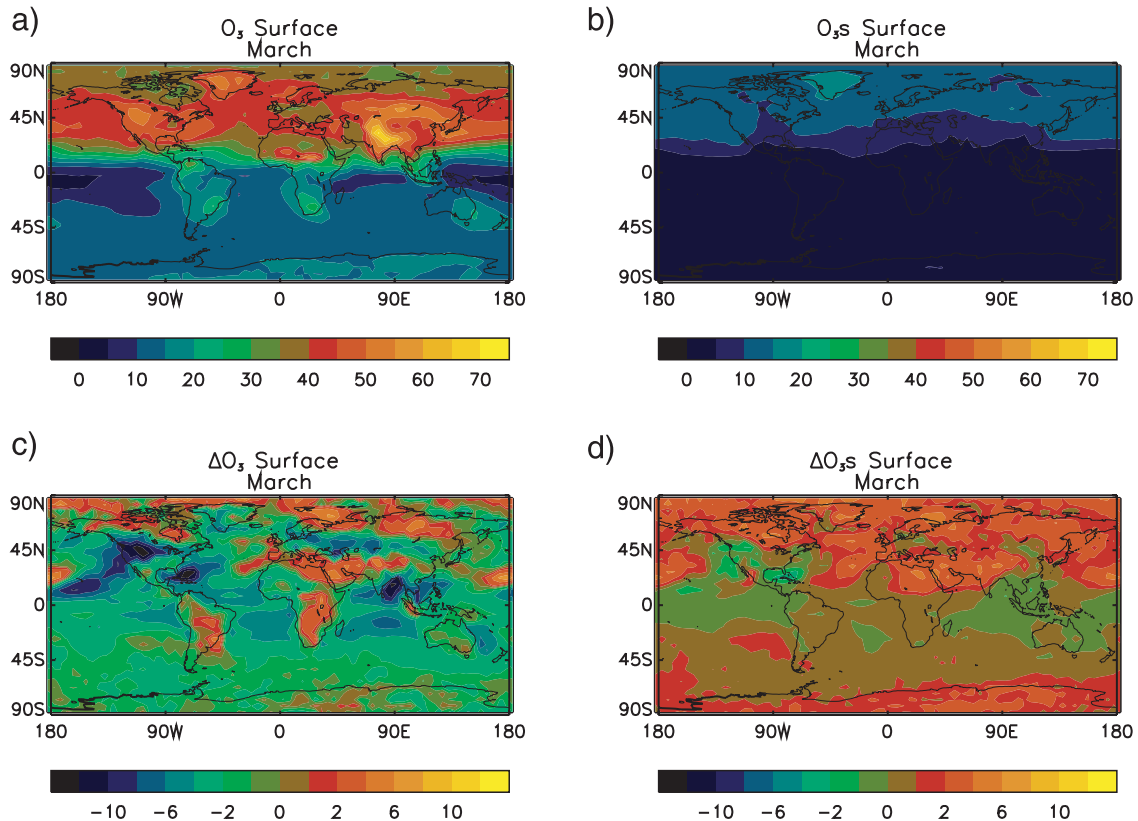
Figure 5. Model surface O_3 concentrations (a) and O_3s concentrations (b), averaged over $1991-1994$. Units are ppb. Surface mean changes between $1991-1994$ and $2091-2094$ in O_3 (c) and O_3s (d). Units are ppb.

the stratospheric ozone chemistry is expected to vary in the future according to changes in the concentrations of important chemical constituents (such as halogens, oxides of nitrogen or methane) and changes in the stratospheric temperature and humidity. The stratospheric circulation obviously also has a large effect on the distribution of ozone. By using a fixed climatology (specified in terms of latitude, longitude and pressure) we are introducing an inconsistency between the stratospheric ozone and the dynamics. This is most evident at the tropopause. The extratropical tropopause pressure decreased by $20-40$ hPa in the 2090s simulation compared to the 1990s. Since the climatological ozone is specified according to absolute pressure rather than relative to the tropopause, the tropopause ozone concentrations are increased in the future run. This variation in tropopause height also means that the stratospheric ozone climatology is imposed over a smaller volume of the model atmosphere in the future.

[17] Figures 4a–4d show the zonal mean concentrations for $1991-1994$, and the fractional differences between the 2090s and the 1990s simulations, for O_3 and O_3s for the month of March averaged over four years. In Figure 4a, a region of upward transport of ozone poor air can be seen in the tropics, with regions of downward transport of ozone rich air in the subtropics and extratropics. The downward transport is particularly noticeable in the northern middle to high latitudes where values of 50 ppb extend down to the lower troposphere. Comparison with Figure 4b shows that $25-50\%$ of this ozone is of stratospheric origin. Figure 4c

shows the fractional change in ozone in the future. There are regions of decreases in concentration around the level of the tropopause in the tropics, high northern latitudes and southern midlatitudes. These are due to a stronger upward flux of ozone poor air (similar decreases are seen in a [7]Be tracer). Decreases in concentration in the lower troposphere, especially the tropics, are caused by the more rapid ozone photochemical loss rate in the more humid climate. There are noticeable regions of ozone increases of between 5 and 15% in the upper troposphere subtropics and southern middle to high latitudes. These are caused by increased downward transport of stratospheric ozone. The increases can be seen more clearly in Figure 4d, which shows the change in O_3s as a fraction of the 1990s full ozone concentration; that is, the denominator is the same as for Figure 4c. The stratospheric ozone component has increased almost everywhere. Although O_3s is subject to the same increase in photochemical loss as the full ozone, the effect is less as O_3s has a shorter residence time in the troposphere.

[18] Figures 5a–5d show the surface concentrations for $1991-1994$, and the differences between the 2090s and the 1990s simulations, for O_3 and O_3s for the month of March averaged over four years. There is a fairly widespread ozone background of $35-45$ ppb over the northern extratropics (Figure 5a). Ozone and its precursors have longer lifetimes in spring compared to the summer, so the marked differences between the continents and the oceans expected in the summertime ozone distributions are not seen here. Particularly noticeable is the lack of a photochemical ozone
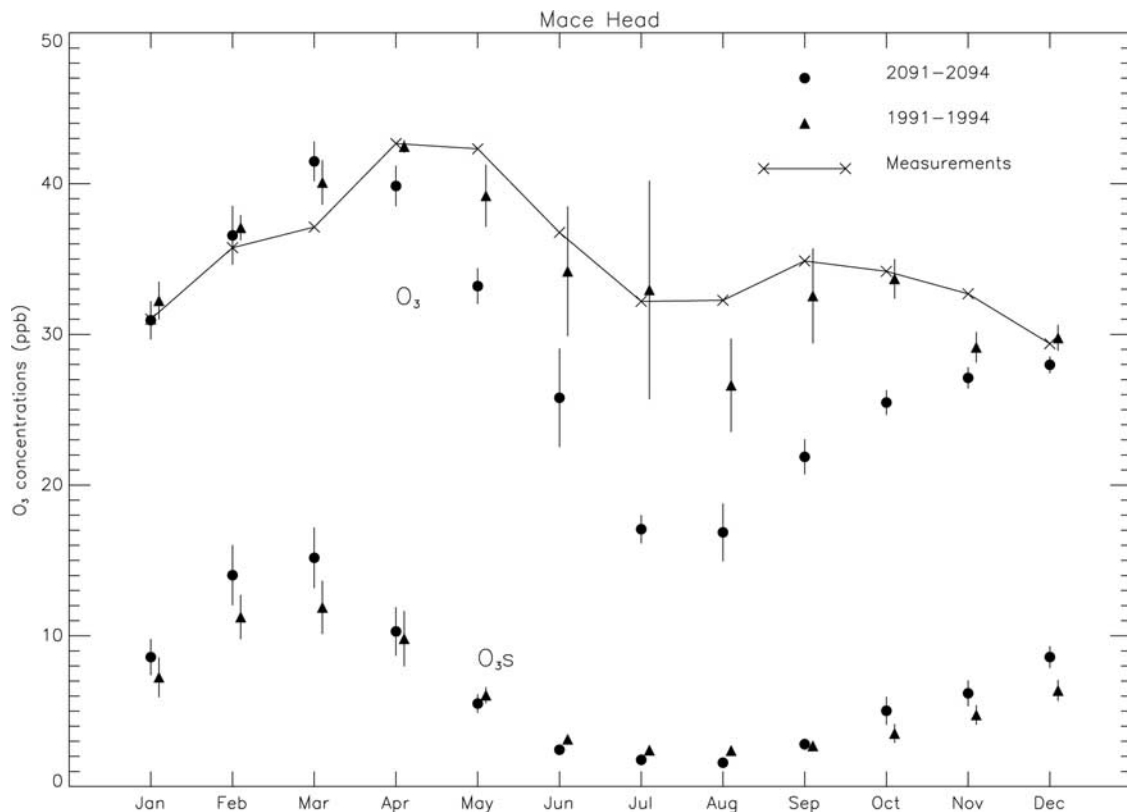
Figure 6. Surface O_3 and O_3s simulated ozone at Mace Head averaged over 1991–1994 (triangles) and 2091–2094 (circles). Measured values are shown as crosses and are taken from *Oltmans and Levy* [1994].

production plume over Europe in March. Ozone from the stratosphere contributes about 10–15 ppb to the background (Figure 5b). Although surface ozone generally decreases in the 2090s simulation, there are localized regions of increased surface ozone in the future in the middle to high northern latitudes (Figure 5c). These correlate well with increases in O_3s (Figure 5d) suggesting that they are due either to increased transport from the stratosphere or by some other process that increases the lifetimes of both O_3 and O_3s. In fact over parts of East Africa, the Arabian Peninsula and northern India the driving meteorology predicts local decreases in humidity in the future and it is likely that this explains the increases in O_3 and O_3s over those locations. The future ozone increases over northern Scandinavia, northern Siberia and the extreme west of Europe are not associated with a drying climate so the correlation with stratospheric ozone increases implies an increase in transport from the stratosphere. The exact location of these regional ozone increases varies when intercomparing individual years in the present and future. However the general pattern of increases in the extreme north and extreme west of Europe is a robust feature. Surface O_3s generally increases in the 2090s simulation in spite of the higher humidity. The areas of large decreases over western America and the Caribbean are associated with particularly high relative increases in the water vapor concentration.

[19] An indication of the effects of future changes in climate is given by Figure 6, showing the current and future seasonal cycles of ozone at Mace Head. The most noticeable signal is a large decrease in summertime ozone in the future due to more efficient ozone loss in the wetter climate. There is an increase in O_3s in the spring due to transport from the stratosphere. This does lead to a slight increase in the full ozone in March although the increase is not statistically significant. The overall effect is for Mace Head surface ozone to peak slightly earlier in the year in future with approximately the same peak concentration but a much lower minimum.

[20] The effect of climate change is summarized in Table 1, which lists the ozone fluxes across 200 hPa, the total loss rate and the ozone burden. The 200 hPa fluxes are listed rather than the tropopause fluxes since the height of the tropopause varies in the two simulations, making a comparison difficult. The net downward flux is larger than the

Table 1. Ozone Transport and Loss Fluxes Averaged Over 4 Years[a]

	1991–1994	2091–2094	Difference
Flux across 200 hPa			
Downward	8057±48	9899±90	1842±102
Upward	6989±45	8433±82	1444±94
Net downward	1067±8	1465±13	398±15
Loss rate	4914±16	5304±14	390±21
Burden (below 200 hPa)	284±2	271±1	−14±2

[a]Fluxes are given in Tg yr^{-1}. Average ozone burden below 200 hPa (Tg). Quoted errors are due to interannual variability only.

range of cross tropopause fluxes given by *Collins et al.* [2000] since it includes some transport within the stratosphere in the high latitudes. The net flux is the difference of two much larger terms, both of which increase in the future climate suggesting that not only is there an increase in the overall transport from the stratosphere by around 37%, but there is also an increase in the two-way exchange of air across the tropopause. The values in Table 1 are annually and globally averaged. There is some seasonal and hemispheric variation as the magnitudes of the transport fluxes are largest in the wintertime of each hemisphere. However, there is still an increase in the net downward ozone flux in each hemisphere and in all seasons.

[21] Since ozone precursor emissions are kept fixed, there is little change in the ozone production rate and all the increase in downward transport is balanced by an increase in loss. The loss rate can increase despite a decrease in the ozone burden due to the shorter ozone lifetime in a more humid atmosphere. If the ozone lifetime could be constrained to remain constant in the future, a rough estimate of the future ozone burden can be obtained by scaling the burden with the necessary increase in loss. This would give a theoretical future ozone burden, due to circulation changes and assuming no change in humidity, of 307 Tg, an increase of 23 Tg or 8%. This will be an overestimate since the largest changes in loss rate occur in the lower troposphere while the bulk of the ozone burden is in the upper troposphere.

4. Conclusions

[22] We have presented an investigation into the effects of circulation changes due to climate change on the stratosphere-troposphere exchange of ozone, and the stratospheric contribution to tropospheric ozone levels. We have tried to do this by holding as many other variables constant as possible. For some variables this is not entirely realistic. For instance the distribution of stratospheric ozone will be strongly affected by changes in the stratospheric circulation. To address this issue rigorously, a full stratosphere-troposphere chemistry model would be needed. Such models are currently under development.

[23] We have identified an increase in the net transport of ozone into the troposphere in the future climate, although a decreased ozone lifetime means that the overall tropospheric burden decreases. There are regions in the midlatitudes to high latitudes where surface ozone is predicted to increase in the spring. However these increases are not significant. Significant ozone increases are predicted in regions of the upper troposphere, suggesting that trends in stratosphere-troposphere exchange should be taken into account when studying the impacts of climate change on radiative forcing. The general increase in the stratospheric contribution (O_3s tracer) to tropospheric ozone in the climate changed scenario indicates that the stratosphere will play an even more significant role in the future.

[24] We have not attempted to identify the physical causes of the predicted increased flux of ozone into the troposphere. In particular the relative importance of the Brewer Dobson circulation, synoptic-scale or convective-scale features. It is doubtful that the GCM in this configuration can accurately represent the Rossby wave action in the midstratosphere as most of the 38 levels are below the tropopause. There are questions too as to how accurately tropopause folds and frontal transport can be represented with a grid size of $3.75° \times 2.5°$.

[25] While this study cannot yet provide a definitive answer on the future changes in the exchange of ozone across the tropopause, it is in qualitative agreement with other studies [*Land and Feichter*, 2003; *Butchart and Scaife*, 2001; *Rind et al.*, 2001]. It suggests that the changes will be significant and that more detailed research into this topic is required. This research will need middle atmosphere models with full treatment of the ozone chemistry of both the troposphere and stratosphere.

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