

## Effects of stratosphere-troposphere chemistry coupling on tropospheric ozone

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[1] A new, computationally efficient coupled stratosphere-troposphere chemistry-climate model (S/T-CCM) has been developed based on three well-documented components: a 64-level general circulation model from the UK Met Office Unified Model, the tropospheric chemistry transport model (STOCHEM), and the UMSLIMCAT stratospheric chemistry module. This newly developed S/T-CCM has been evaluated with various observations, and it shows good performance in simulating important chemical species and their interdependence in both the troposphere and stratosphere. The modeled total column ozone agrees well with Total Ozone Mapping Spectrometer observations. Modeled ozone profiles in the upper troposphere and lower stratosphere are significantly improved compared to runs with the stratospheric chemistry and tropospheric chemistry models alone, and they are in good agreement with Michelson Interferometer for Passive Atmospheric Sounding satellite ozone profiles. The observed CO tape recorder is also successfully captured by the new CCM, and ozone-CO correlations are in accordance with Atmospheric Chemistry Experiment observations. However, because of limitations in vertical resolution, intrusion of CO-rich air in the stratosphere from the mesosphere could not be simulated in the current version of S/T-CCM. Additionally, the simulated stratosphere-to-troposphere ozone flux, which controls upper tropospheric OH and O<sub>3</sub> concentrations, is found to be more realistic in the new coupled model compared to STOCHEM.

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### 1. Introduction

[2] Changes in trace gas emissions caused by human activity have a profound impact on ozone (O<sub>3</sub>) in both the stratosphere and troposphere. During the past few years, a number of chemistry-climate models (CCMs) have been developed to understand and quantify the influence of the key chemical and physical processes that determine ozone concentrations. Such an understanding enables us to predict future ozone evolution, particularly in the stratosphere, in changing climate [e.g., Austin *et al.*, 2003; Rotman *et al.*, 2004; Jöckel *et al.*, 2006; Eyring *et al.*, 2006, 2007, and

references therein]. With the help of such CCMs, a better understanding has been obtained with regard to past and future changes of total column ozone, and results suggest that stratospheric ozone is likely to return to its pre-1980 state by the mid-21st century [e.g., *World Meteorological Organization (WMO)*, 2007]. Future projections of tropospheric ozone are still uncertain and are strongly dependent on regionally variable ozone precursor emission scenarios and climate change [e.g., Stevenson *et al.*, 2006]. Some CCMs and chemical transport models (CTMs) have also been used to understand the effects of emissions and various climate processes on tropospheric ozone abundances both on a regional and global scale [e.g., Stevenson *et al.*, 2006; Wild, 2007; Zeng *et al.*, 2008; Wu *et al.*, 2007]. Most of these model studies indicate that tropospheric ozone has increased significantly since preindustrial times [e.g., Gauss *et al.*, 2006], but key processes and underlying mechanisms controlling the tropospheric ozone budget are still poorly constrained, making future projections uncertain [Stevenson *et al.*, 2006; Wild, 2007].

[3] Despite the rapid development of CCMs in recent years, the CCMs/CTMs used in previous tropospheric studies differ in various aspects, and hence they show large differences and uncertainties in modeled lower atmosphere ozone [Wild,

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2007]. For instance, most of these CCM/CTMs have only tropospheric chemistry included with a fixed stratospheric influx of ozone at the model's top chemistry level, while some models relax or nudge toward an ozone climatology above a certain level. This inevitably introduces biases in the predicted tropospheric ozone as the ozone input via stratosphere-troposphere exchange (STE) will vary with time/climate. Previous studies have shown convincing evidence that STE is one of the most important sources of tropospheric ozone [e.g., Gettelman et al., 1997; Collins et al., 2003; Roelofs et al., 2003; Olsen et al., 2004; Stevenson et al., 2006]. Some other CCMs/CTMs have only considered chemistry and transport processes in the model without the coupling of the radiatively active chemical species from the chemistry scheme to the model's radiation schemes.

[4] Among various things, a reasonable time-dependent stratospheric input of some key chemical species including  $O_3$  and  $HNO_3$  is crucial for better prediction of tropospheric ozone, particularly in the upper troposphere. However, most existing CCMs typically have either detailed tropospheric or stratospheric chemistry schemes, but rarely both. This is one of the most important missing mechanisms in the model, which limits accurate simulation of the tropospheric ozone concentrations.

[5] In the development of CCMs, the combination of different well-tested submodels into a single framework is found to be an efficient way to set up whole atmospheric CCMs. Two examples include the ECHAM/MESSEy Atmospheric Chemistry model [Jöckel et al., 2006] and the IMPACT model [Rotman et al., 2004], which both include detailed descriptions of tropospheric and middle atmosphere chemistry. In this study, we set up a CCM with three mature submodels including a well-documented general circulation model (GCM) and two chemistry modules for stratospheric and tropospheric chemical processes (hereafter referred as S/T-CCM). The model setup and numerical experiments are described in section 2. The S/T-CCM-simulated  $O_3$ , CO,  $CH_4$ , and  $HNO_3$  are evaluated for present-day conditions against various satellite and ozonesonde data sets in section 3. We also investigate the impact of coupling on predictions of tropospheric chemistry, which is discussed in section 4. In a companion paper by R. Damoah et al. (Tropospheric ozone response to climate change: Results from a new stratosphere/troposphere chemistry-climate model, submitted to *Atmospheric Chemistry and Physics Discussion*, 2010), the CCM is used to examine potential future changes in tropospheric chemistry related to climate change. Our summary and conclusions are presented in section 5.

## 2. Coupled Chemistry-Climate Model

[6] Our new CCM consists of three components: a GCM, a stratospheric chemistry module, and a tropospheric chemistry module. The GCM is based on the troposphere-stratosphere version of the UK Met Office Unified Model (UM) [Cullen, 1993; Swinbank et al., 1998; Pope et al., 2000]. The UM has a horizontal resolution of  $2.5^\circ \times 3.75^\circ$  and 64 levels extending from the surface to 0.01 hPa ( $\sim 80$  km). In a previous study by Tian and Chipperfield [2005], this GCM was coupled with a stratosphere chemistry module from the SLIMCAT CTM [Chipperfield, 1999] to study ozone-climate interactions. Subsequent long-term climate simulations using this model

(named UMSLIMCAT) show that both the GCM and the stratospheric chemistry module have a good performance in simulating the stratospheric dynamics and chemistry [Waugh and Eyring, 2008]. UMSLIMCAT participated extensively in the Stratospheric Processes and their Role in Climate (SPARC) CCMVal-1 and CCMVal-2 intercomparisons. In the current S/T-CCM version, we have included an updated SLIMCAT CTM chemistry scheme combined with a tropospheric chemistry transport model (STOCHEM) [Stevenson et al., 2004], and hence the model calculates chemistry from the surface up to 0.5 hPa.

### 2.1. Stratospheric Chemistry

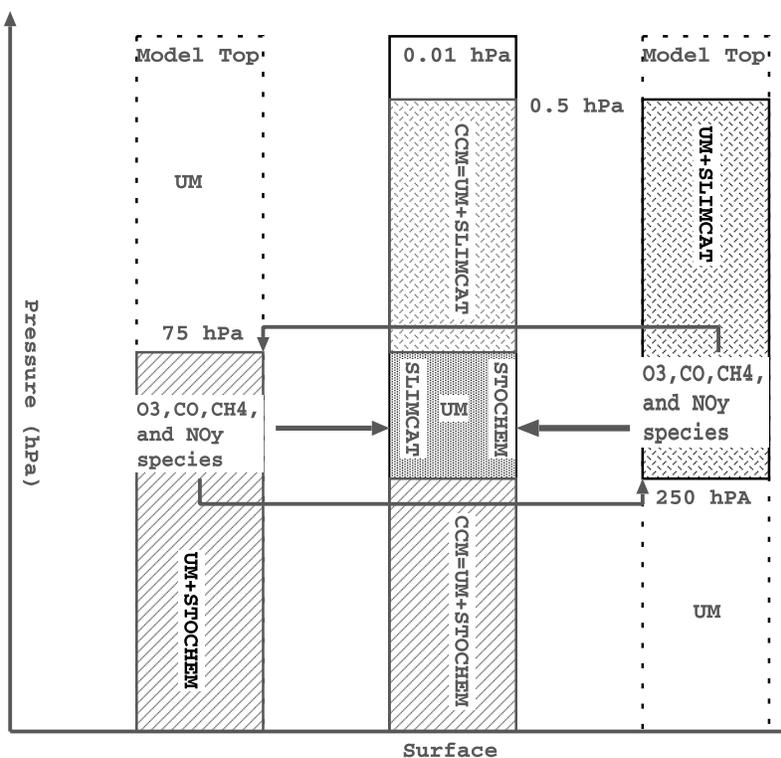
[7] As mentioned above, the stratospheric chemistry module is adopted from the SLIMCAT CTM [Chipperfield, 1999, 2006], and it contains gas-phase chemistry of  $O_x$ ,  $HO_x$ ,  $NO_y$ ,  $Cl_y$ , and  $Br_y$  species, long-lived source gases, and a  $CH_4$  oxidation scheme. Heterogeneous chemistry is calculated on sulphate aerosols and on polar stratospheric clouds (PSCs). The PSC scheme is based on an equilibrium approach with gas-phase species (e.g.,  $H_2O$ ,  $HNO_3$ ) being condensed onto aerosol particles with a specified number density distribution. Stratospheric sulphate aerosol surface area densities come from a climatology derived from satellite observations [SPARC, 2006].

[8] The stratospheric chemistry module uses 28 advected tracers, and the  $CH_4$ ,  $N_2O$ ,  $H_2O$ , and  $O_3$  fields are coupled to the UM's radiation scheme above 250 hPa. From 250 to 0.5 hPa, the chemistry is calculated by the SLIMCAT CTM chemistry scheme at all grid points. Those 28 tracers are advected in the UM using the QUINTIC-MONO scheme [Gregory and West, 2002]. This scheme performs much better than others in maintaining realistic tracer distributions [Tian and Chipperfield, 2005].

[9] The chemistry is not calculated above 0.5 hPa for the purpose of saving computer time. There are no tracer upper boundary conditions at 0.5 hPa either. The tracers above 0.5 hPa are passively advected, whereas below 0.5 hPa, they are calculated by UMSLIMCAT. This has little influence on chemistry processes within the upper troposphere and lower stratosphere (UTLS) region.

### 2.2. Tropospheric Chemistry

[10] From the surface to 75 hPa, chemistry is calculated by STOCHEM, and the chemical species are advected with a Lagrangian scheme driven by the UM's meteorological fields. STOCHEM was first described by Collins et al. [1997]; the version used here incorporates major updates to the chemistry [Collins et al., 1999], convection [Collins et al., 2002], and surface exchange [Sanderson et al., 2003]. STOCHEM uses a Lagrangian transport scheme, dividing the atmosphere into 50,000 equal mass air parcels, which are then advected by UM wind fields, supplied at 3 hourly intervals. A chemistry solver operates within each air parcel, and there are parameterizations for interparcel mixing and convection. STOCHEM has 70 chemical species, including 11 nonmethane hydrocarbons (including isoprene), a detailed description of the sulfur cycle, and aqueous-phase chemistry. Halogens are not included in STOCHEM, and heterogeneous chemistry in the model is limited to the reaction of  $N_2O_5$  on aerosol [e.g., Dentener and Crutzen, 1993; Jacob, 2000]. The model sulphate aerosol distribution is used for this. The



**Figure 1.** Details of the coupling between the tropospheric (STOCHEM) and stratospheric (UMSLIMCAT) chemistry schemes in the S/T-CCM.

model is highly efficient and produces results comparable to similarly sophisticated Eulerian models [e.g., Dentener *et al.*, 2005; Stevenson *et al.*, 2006].

### 2.3. Coupling of Chemical Schemes

[11] Figure 1 illustrates the details of the coupling of the two chemical schemes. In the coupled S/T-CCM, the chemistry scheme from SLIMCAT is integrated into the UM from 250 to 0.5 hPa, while the STOCHEM chemistry scheme is used from the surface to 75 hPa. The two chemistry schemes are kept separate and run in parallel. There is an overlapping region (75–250 hPa) where both schemes operate and the overall CCM profile is a merged combination of the results of the two schemes. Chemical fields are passed between the two chemical modules every 30 min time step at the point that the separate chemical calculations are performed. At the lower boundary of the stratospheric chemistry (250 hPa), relevant fields are overwritten by values from STOCHEM for that level. At pressures of <75 hPa, the stratospheric values are left unchanged. Similarly, at the upper boundary of STOCHEM (75 hPa), values are overwritten by values from the stratospheric scheme for that level. In the coupled S/T-CCM, the ozone column used for photolysis rates in STOCHEM comes from the SLIMCAT chemistry scheme, whereas in the uncoupled version of STOCHEM, an  $O_3$  climatology was used.

[12] Between 250 and 75 hPa, certain species common to both schemes ( $O_3$ ,  $CH_4$ ,  $CO$ , and the  $NO_y$  species) are overwritten in both submodels by merged values, using a weighting function that varies linearly with pressure between 75 and 250 hPa. Note that the number of the merged tracers

can be increased as required as the respective chemical schemes develop. The weighting is actually given to the two chemical schemes SLIMCAT and STOCHEM, rather than to stratosphere and troposphere. The basis for this is that STOCHEM has more reasonable tracer predictions below 150 hPa, no matter if 150 hPa is in the stratosphere or in the troposphere, while SLIMCAT has more reasonable predictions above 150 hPa (see section 3). Therefore, more weighting is given to SLIMCAT-predicted values above 150 hPa and to STOCHEM-predicted values below 150 hPa without considering whether these layers are in the troposphere or stratosphere. The coupled model improves on both the separate STOCHEM and UMSLIMCAT models by providing more realistic upper/lower boundary conditions. This method of combining the two submodels does not implicitly conserve mass of individual species. However, analysis of time series for key species in the UTLS region indicates that they rapidly equilibrate between the two submodels and there are no significant trends, suggesting that the hybrid model methodology used here is justified.

[13] Using these two well-tested chemistry schemes together to set up a whole atmosphere CCM is a more efficient approach to CCM development than developing an integrated whole atmosphere chemistry scheme that can be performed throughout all GCM levels. Extending STOCHEM upward on all model levels makes simulation effectively impossible as STOCHEM is based on constant equal mass air parcels (in the stratosphere, these would occupy very large volumes). On the other hand, operating the Eulerian scheme of the SLIMCAT kind with a more complex chemistry scheme throughout all GCM levels would significantly increase

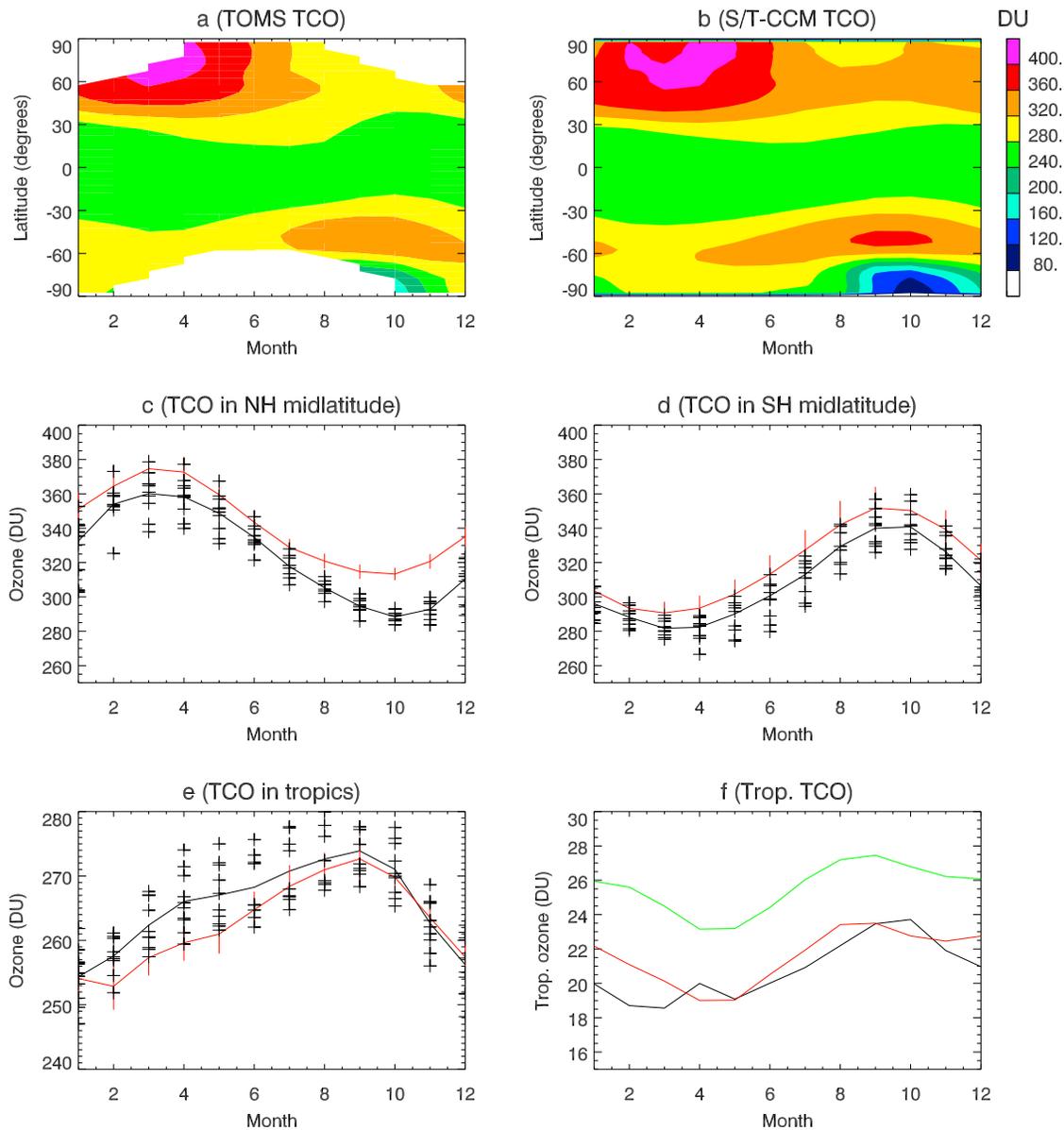
**Table 1.** Chemistry Schemes Used in Model Experiments

Experiment	Configuration
COUPLED	Coupled stratospheric and tropospheric chemistry in the UM
STRAT	Stratospheric chemistry (SLIMCAT) in the UM
TROP	Tropospheric chemistry (STOCHEM) in the UM

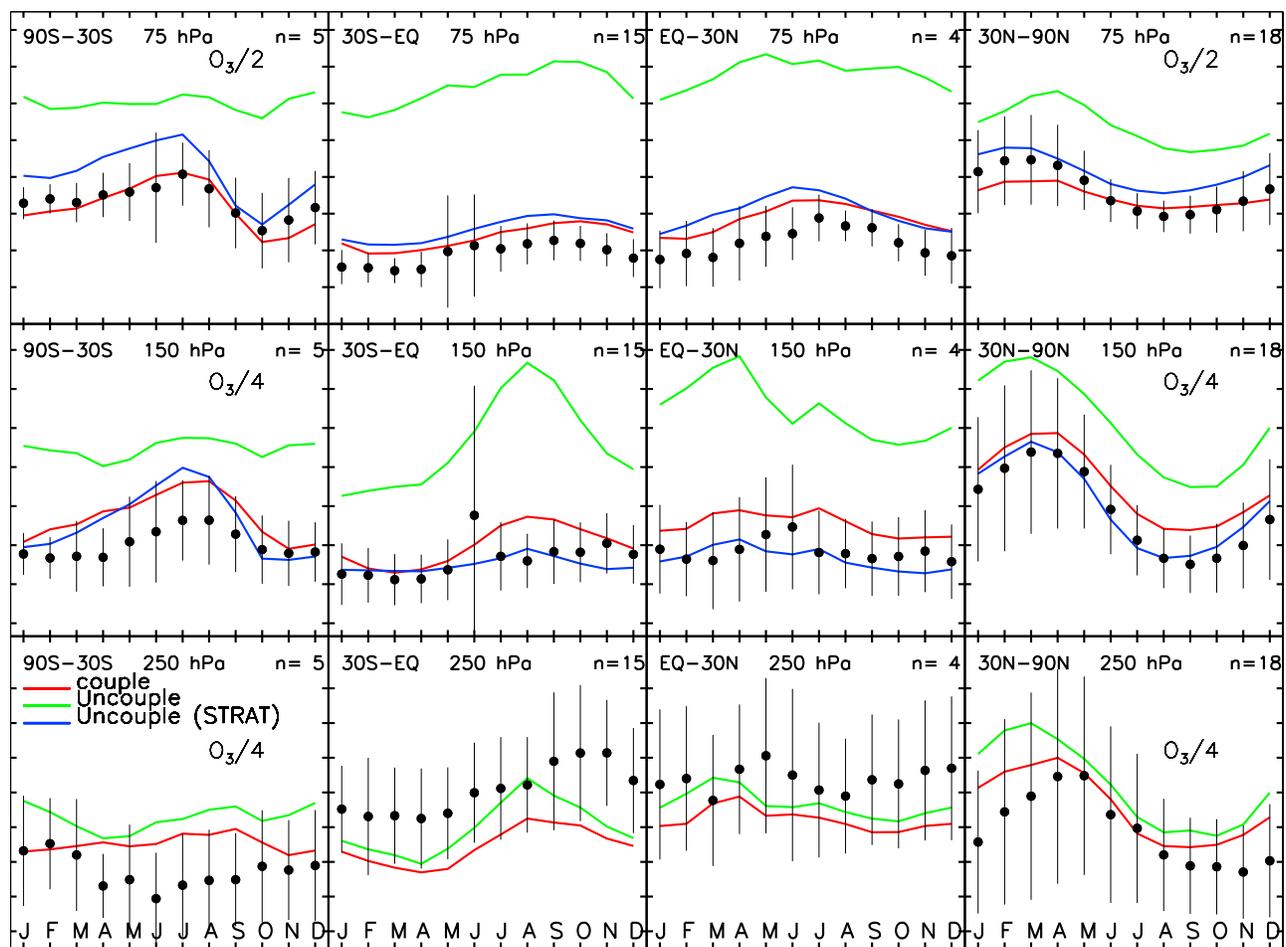
computer time compared with the CCM described in this study. This increase in CPU time depends on the level of sophistication of the tropospheric chemistry scheme. Compared to the cost of the previous separate CCMs (UMSLIMCAT and STOCHEM), which were already computationally efficient, the additional chemistry adds about 50% to the total cost of the model. This is only a modest increase given the complexity of the resulting S/T-CCM.

## 2.4. Model Experiments

[14] Three experiments have been performed for this study (see Table 1). In the control run, the two chemistry modules are coupled together within the UM (hereafter referred to as



**Figure 2.** Zonal mean total column ozone (DU) climatologies from (a) TOMS observations for the period from 1989 to 1996 and (b) run COUPLED. The annual cycle of the monthly mean TCO averaged over (c) 30°–60°N, (d) 30°–60°S, and (e) 30°–30°N from run COUPLED (red line) and TOMS (black line) are shown with their corresponding interannual variabilities (bars and pluses). (f) Annual cycle of the monthly mean tropospheric ozone column from the TOMS observations (black line), run COUPLED (red line), and run TROP (green line).



**Figure 3.** Comparison of the annual cycle of monthly mean ozone observations (black dots) and model runs TROP (green), STRAT (blue), and COUPLED (red) sampled for different latitude bands ( $90^{\circ}$ – $30^{\circ}$ S,  $30^{\circ}$ S to equator, equator to  $30^{\circ}$ N, and  $30^{\circ}$ – $90^{\circ}$ N) and different pressure levels (75, 150, and 250 hPa). Each panel is the mean of many years from several sites (the number of sites is given in the top right corner of each plot); the model and observations are sampled in the same way. The bars for each observation are the average of the interannual standard deviations at each station. Data are from Logan [1999] and Thompson *et al.* [2003a, 2003b].

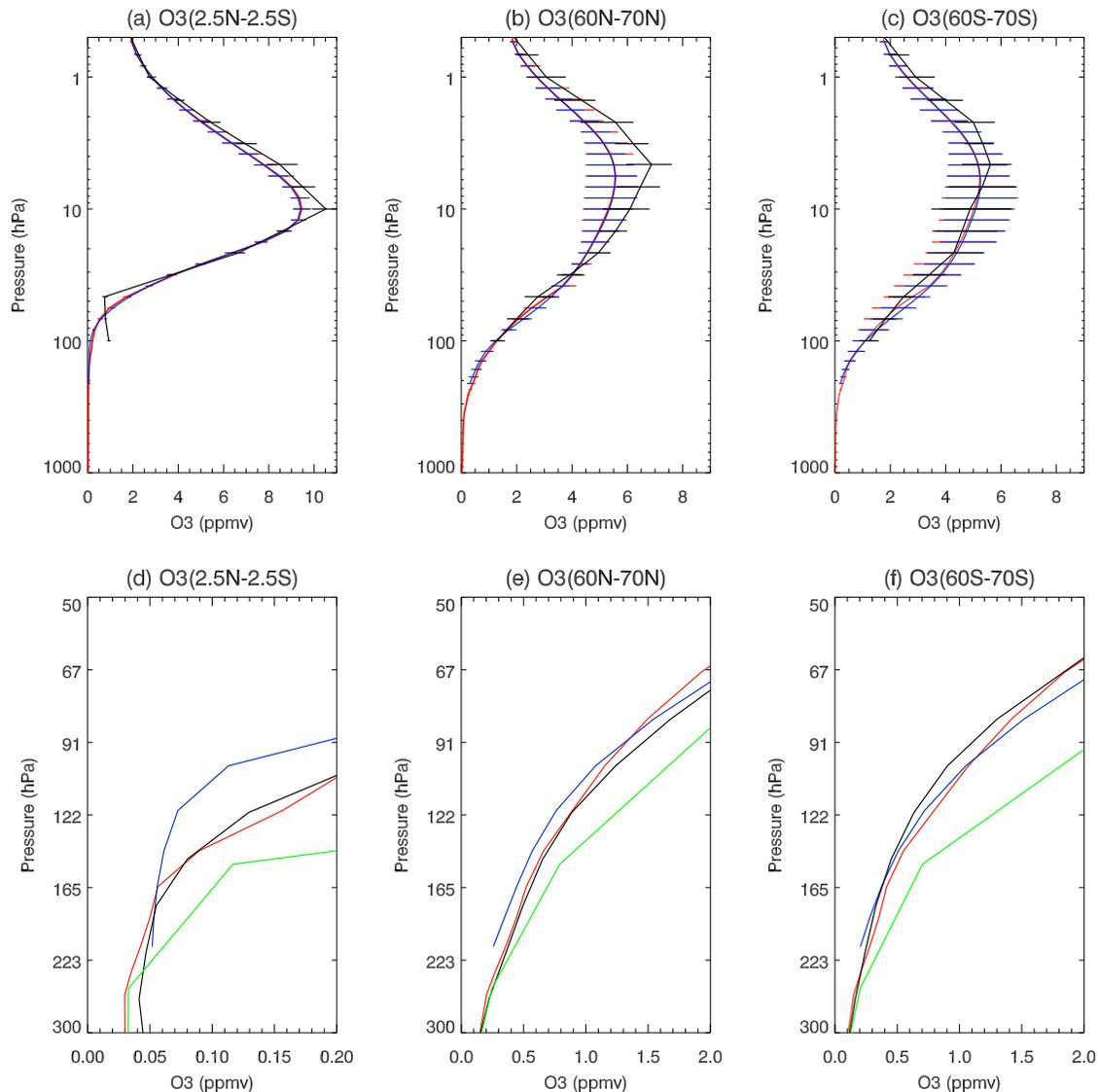
COUPLED). In runs STRAT and TROP, the stratospheric and tropospheric chemistry are calculated separately as in the respective older versions of the UM (hereafter “uncoupled runs”). The run STRAT uses specified surface mixing ratios of source gases as its lower boundary condition. These tracers are passive below the lowest level at which stratospheric chemistry is calculated (250 hPa). Overwriting the surface mixing ratio of chemical families acts as the sink/source for these species. In run TROP (STOCHEM), air parcels above the tropopause relax their ozone concentrations toward the ozone climatology with a 10 day relaxation time scale [Li and Shine, 1995]. All other model configurations for the three simulations are the same.

[15] All runs are for 11 years and use a monthly varying but annually invariant sea surface temperature (SST) and sea-ice climatology that is based on average values during the period 1995–2005, from the SST and sea-ice fields used by Eyring *et al.* [2006]. Greenhouse gas values including  $\text{CO}_2$  and chlorofluorocarbons for 1990 are taken from the Intergovernmental Panel on Climate Change scenario [WMO,

2003]. Tropospheric methane is initialized to 1760 ppbv, which is fixed in run STRAT and is in approximate equilibrium with surface emissions in runs COUPLED and TROP. Surface emission fields for all other trace gases are as described by Stevenson *et al.* [2006]. The first year of data is considered as spin-up and is excluded from the analysis. All model results presented here are 10 year averaged fields, except where stated otherwise. Climate fields were initialized from dumps taken from multidecadal runs of a coupled ocean-atmosphere climate model without interactive chemistry and are considered to be close to equilibrium, which therefore allows only a short spin-up time. Chemical species in the stratosphere are initialized from multiyear real-time runs of the SLIMCAT/TOMCAT chemistry transport model [Chipperfield, 2006] that are also close to steady state conditions.

### 3. Model Climatology Versus Observations

[16] We noted above that the previous UM versions with only either the stratospheric chemistry or the tropospheric

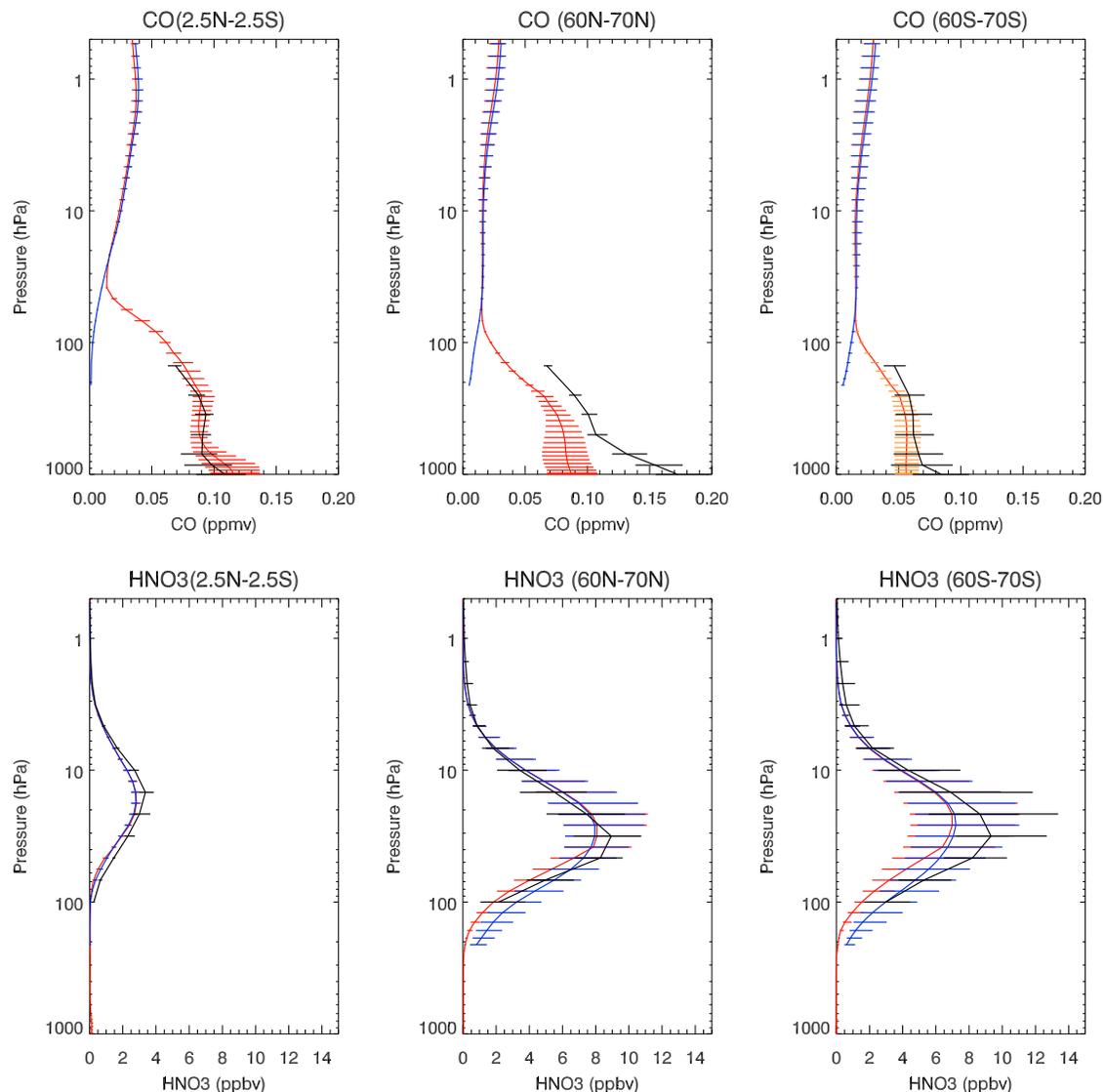


**Figure 4.** (a–c) Annual mean profiles of O<sub>3</sub> from runs COUPLED (red line) and STRAT (blue line) averaged over different latitude bands. The horizontal bars show the minimum and maximum range of the monthly mean values. The corresponding profiles from the UARS climatologies (black line) are also shown for comparison. (d–f) Annual mean profiles of O<sub>3</sub> within the UTLS region from runs COUPLED (red line), STRAT (blue line), and TROP (green line) averaged over three different latitude bands. The corresponding profiles from the MIPAS climatologies for the period 2002–2009 (black line) are also shown for comparison.

chemistry have performed well in simulating both dynamics and chemistry [e.g. Tian and Chipperfield, 2005; Stevenson *et al.*, 2006]. This is one reason why we want to explore ways in which these models can be improved, without the cost of creating a new whole atmosphere CCM. Here we will focus on how the coupling of the two chemical regions improves the performance over the former uncoupled CCMs.

[17] Figure 2 shows modeled total column ozone from run COUPLED and Total Ozone Mapping Spectrometer (TOMS) observations. It is apparent that the total column ozone (TCO) from run COUPLED is in good agreement with TOMS both in spatial distribution and annual cycle, which is due, in part, to the realistic tropospheric contribu-

tion. The TCO values at middle latitudes in both hemispheres from run COUPLED are slightly higher (~10 DU) than the TOMS observations (Figures 2c and 2d), while in the tropics, the modeled TCO is slightly lower (Figure 2e). However, these discrepancies are well within the range of 1 standard deviation from the mean TCO climatologies. Figure 2f further shows seasonal variations of the partial ozone column in the tropics (averaged between 12°S and 12°N) from runs COUPLED and TROP. The observed tropical tropospheric column ozone, derived from TOMS data using the convective cloud differential method [Ziemke *et al.*, 1998], is also shown for comparison. We can see that the coupling of two chemistry schemes improves the simulation of the tropical tropospheric column ozone compared



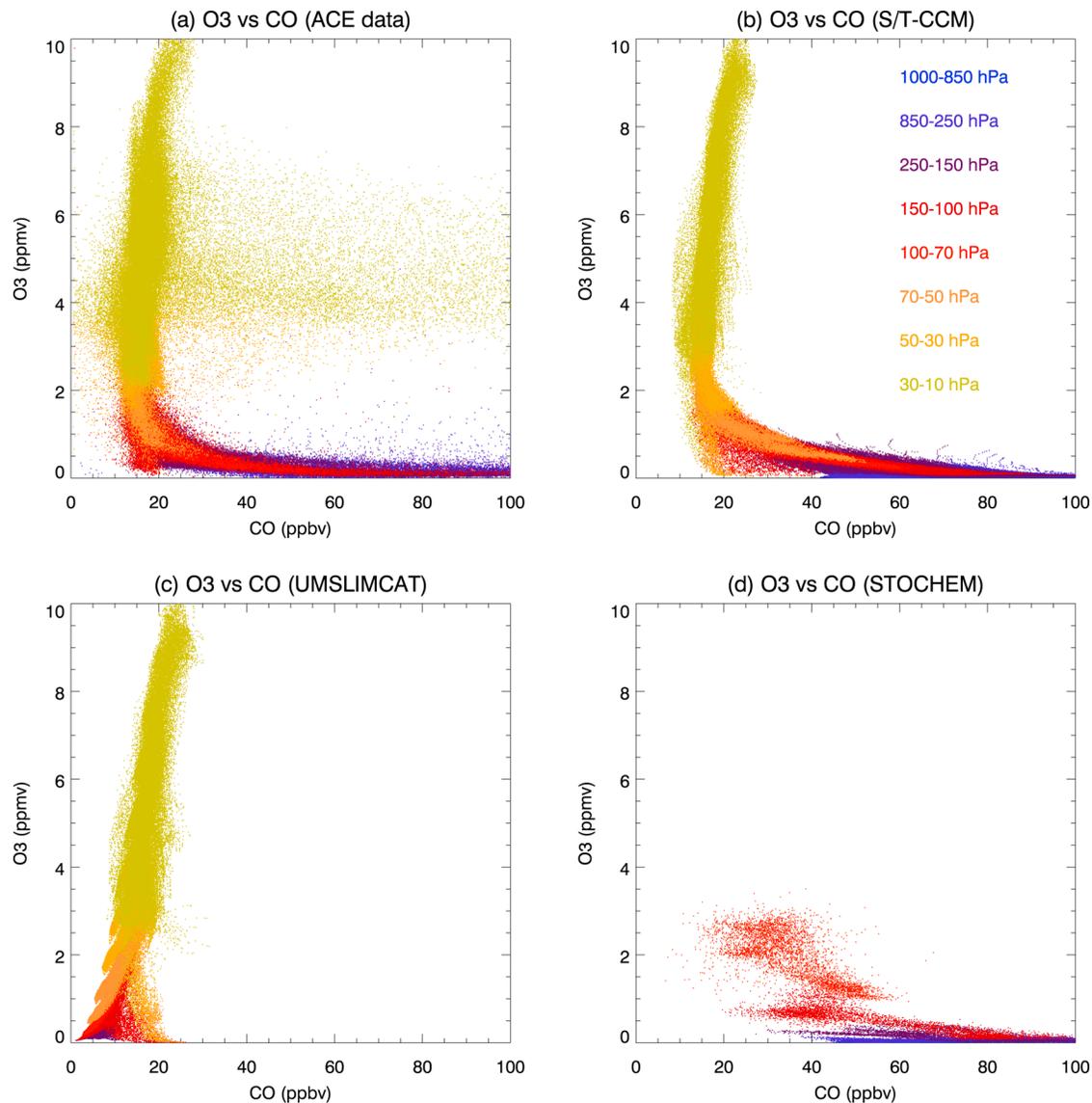
**Figure 5.** Annual mean profiles of CO (top) and HNO<sub>3</sub> (bottom) from the runs COUPLED (red line) and STRAT (blue line) averaged over different latitude bands. The horizontal bars show the minimum and maximum range of the monthly mean values. Corresponding profiles from the MOPITT CO observations and UARS HNO<sub>3</sub> climatology (black lines) are shown for comparison.

to run TROP. It is apparent that coupling improves the ozone predictions within the UTLS, resulting in a more reasonable ozone upper boundary condition for STOCHEM; consequently, the tropospheric TCO is improved. Note that we cannot reasonably expect the model to capture the interannual variability of the TCO because of the fixed SST climatology and other parameters in the model configuration. Figures 2c–2e clearly show that the interannual variability of the TCO in S/T-CCM is much less than that of the TOMS observations.

[18] Figure 3 shows the annual cycle of monthly mean ozone from sonde observations and model runs COUPLED, TROP, and STRAT, sampled for different latitude bands and pressure levels in the UTLS. The ozone predictions from the coupled model S/T-CCM are overall in accordance with those in the uncoupled STRAT run above 150 hPa, and coupling of the tropospheric chemistry improves ozone

predictions at 75 hPa relative to those in run STRAT. It is apparent that at 75 and 150 hPa, ozone is strongly overestimated in run TROP. In the high-latitude upper troposphere (i.e., 250 hPa), ozone is also improved in the coupled run relative to those in run TROP. At lower latitudes, there seems to be no significant improvement in the ozone predictions at 250 hPa compared to sonde observations. However, it will be shown later that the simulated ozone by coupled model S/T-CCM is overall improved within the UTLS region at all latitudes.

[19] The modeled annual mean O<sub>3</sub> profiles from the surface to 0.5 hPa averaged over different latitude bands from runs STRAT and COUPLED are shown in Figures 4a–4c. The corresponding profiles derived from UARS measurements provided by the SPARC Data Center (<http://www.sparc.sunysb.edu/html/randel.html>) are also plotted. O<sub>3</sub> values in runs COUPLED and STRAT are very similar in



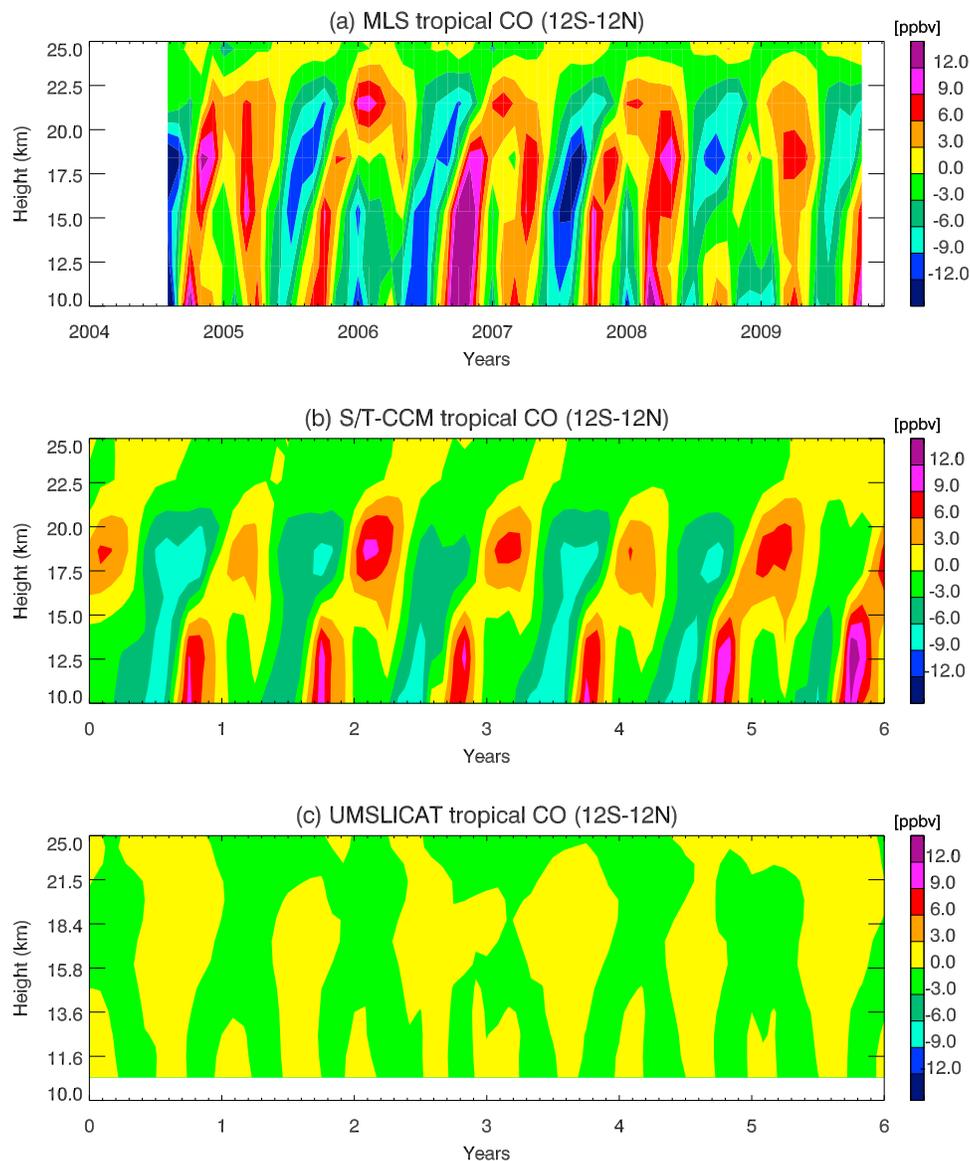
**Figure 6.** CO versus O<sub>3</sub> correlations based on the data from (a) ACE observations, (b) run COUPLED, (c) run STRAT, and (d) run TROP. Different colors represent data points at different levels. The ACE data points plotted are instantaneous values of O<sub>3</sub> and CO at all latitudes where data are available, whereas the model values are monthly means at the same locations.

the upper stratosphere, indicating that the coupling of two chemistry schemes has no significant effect on the annual mean ozone values above ~50 hPa. Note also that the modeled O<sub>3</sub> values in the northern high-latitude upper stratosphere are slightly lower than the SPARC climatology, but at the southern high latitudes and in tropics, the modeled stratospheric ozone is close to the SPARC climatology.

[20] Figures 4d–4f also show annual mean profiles of O<sub>3</sub> in the UTLS region (from 300 to 50 hPa) from the three runs. The corresponding profiles derived from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) data for the period 2002–2009 are also plotted for comparison. We can see that the UTLS O<sub>3</sub> in the coupled run COUPLED is in good agreement with the MIPAS observations at all selected latitudes. In contrast, the UTLS O<sub>3</sub> is significantly overestimated in run TROP above 250 hPa

but underestimated in run STRAT relative to the MIPAS measurements.

[21] Similar to Figure 4, Figure 5 shows annual mean CO and HNO<sub>3</sub> profiles from runs COUPLED and STRAT. The corresponding HNO<sub>3</sub> profiles derived from the UARS climatology and CO profiles based on the Measurements of Pollution in the Troposphere (MOPITT) from 2002 to 2006 are also plotted for comparison. The CO values simulated by the S/T-CCM are well within the range of the corresponding MOPITT observations in the tropics and southern higher latitudes. At high northern latitudes, modeled CO values are lower than MOPITT data, probably mainly because of inaccuracies in the surface CO emissions [see *Shindell et al.*, 2006]. HNO<sub>3</sub> values in S/T-CCM are slightly smaller than the UARS climatology at all latitudes but are much smaller in the southern high latitudes, possibly because of unrealistic HNO<sub>3</sub> values calculated by STOCHEM.



**Figure 7.** Time variation of tropical CO (averaged between 12°N and 12°S) with the annual average removed (ppbv) from (a) MLS observations, (b) run COUPLED, and (c) run STRAT. Note that only six years of model data are shown since the MLS CO observations cover years from 2004 to 2009.

[22] To further validate modeled CO and O<sub>3</sub> profiles, Figure 6 shows the correlations of modeled monthly mean CO and O<sub>3</sub> values and the Atmospheric Chemistry Experiment (ACE) [Bernath *et al.*, 2005; Clerbaux *et al.*, 2005] CO and O<sub>3</sub> profiles from 2004 to 2005. The modeled zonal mean CO and O<sub>3</sub> are sampled in the same way as those of the ACE data over the selected latitude bands. Note that CO versus O<sub>3</sub> correlations are well captured by the coupled S/T-CCM compared to the ACE data. In the middle stratosphere from 10 to 30 hPa, the ACE CO shows some high values attributed to the descent of CO-rich mesospheric air that is not treated in our S/T-CCM. In addition, the ACE CO and O<sub>3</sub> profiles are instantaneous values, while the modeled CO and O<sub>3</sub> shown are the monthly mean values; consequently, there is larger variability in the ACE CO and O<sub>3</sub> correlation plot. Figures 6c and 6d give the CO and O<sub>3</sub> correlations from the uncoupled runs STRAT and TROP. It is

apparent that CO and O<sub>3</sub> correlations in the lower stratosphere from 100 to 50 hPa are significantly improved in the S/T-CCM compared to those in TROP (i.e., the tropospheric chemistry-only simulation). The results indicate that the coupling of two chemistry schemes significantly improves O<sub>3</sub> and CO in the lower stratosphere. A comparison of Figures 6a and 6c reveals that the coupling of two schemes leads to no significant changes in CO and O<sub>3</sub> correlations in the upper stratosphere. Also noticeable is that CO and O<sub>3</sub> in run STRAT (i.e., the stratospheric chemistry-only simulation) are clearly not well correlated in the UTLS region from 250 to 50 hPa.

#### 4. Modeled CO and Ozone STE Flux

[23] Studies of atmospheric chemistry and transport within the UTLS region are hampered by the lack of high-resolution

**Table 2.** Ten Year Averaged Tropospheric O<sub>3</sub> Budget Terms and CH<sub>4</sub> Lifetimes, With Respect to Oxidation by OH, for Runs COUPLED and TROP

Experiment	P <sup>a</sup>	L	D	$S_{inf}$	$\tau_{CH_4}$ (years)
COUPLED	4596	4185	1030	620	10.85
TROP	4578	4357	1071	850	11.16

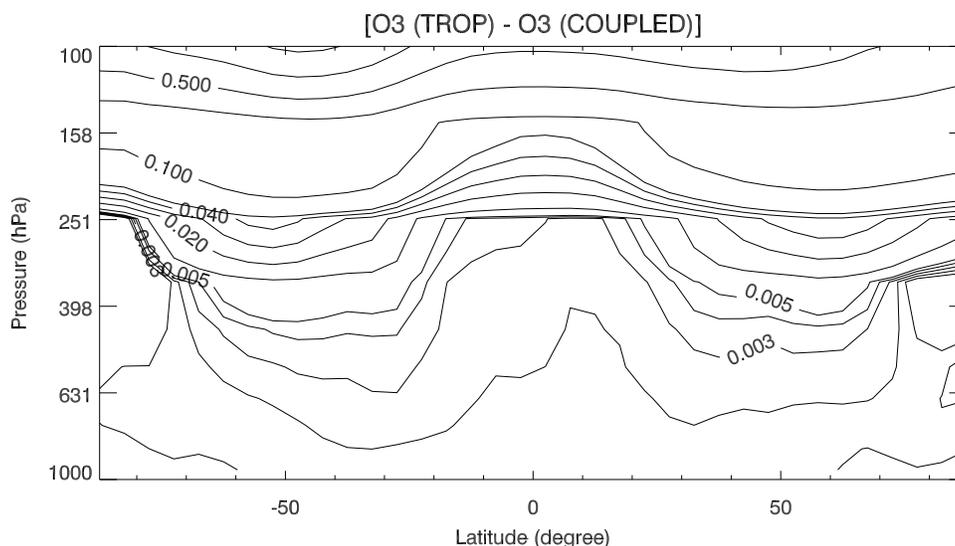
<sup>a</sup>P, chemical production; L, chemical loss; D, surface deposition;  $S_{inf}$ , stratospheric input (inferred from the residual of the other terms (all in Tg (O<sub>3</sub>) yr<sup>-1</sup>));  $\tau_{CH_4}$ , methane lifetime.

observations. An understanding of some processes therefore depends on the availability of detailed models that are evaluated against the observations that are available. Figure 7 shows time variations of the modeled CO in runs COUPLED and STRAT within the tropical UTLS region; also shown are the corresponding CO satellite observations by Microwave Limb Sounder (MLS) from 2004 to 2009. Note that in the COUPLED run, a weak signal of the CO tape recorder can be clearly seen in the tropical UTLS. This kind of CO tape recorder has also been detected in satellite CO observations [e.g., *Schoeberl et al.*, 2006] and is thought to be related to the annually varying mixing ratios of CO in upper tropospheric air transported into the stratosphere because of biomass-burning-related CO emissions. The amplitude of the CO tape recorder in the modeled CO is about 12 ppbv, slightly smaller than that in the satellite CO measurements shown in Figure 7a, which is about 20 ppbv. Note also that the modeled CO tape recorder actually starts from 14 km (around 360 K isentropic surface) and fades out above 20 km, but in the MLS CO, the head of the tape recorder extends slightly higher to 23 km. The result in Figure 7b is overall consistent with the MLS CO data shown in Figure 7a, suggesting the convection and transport processes within the UTLS are reasonable in the coupled S/T-CCM.

[24] There is no CO tape recorder signal in run STRAT as the fixed CO boundary condition in the upper troposphere (mixed from the fixed surface value) cannot generate a realistic CO time variation. It has been argued that the CO tape recorder is more determined by convection and diabatic

uplift within the tropical tropopause layer [*Dessler*, 2002; *Schoeberl et al.*, 2006]. Although run STRAT has nearly the same dynamics as run COUPLED, it does not have the time-varying surface emissions and CO distribution. Therefore, it cannot generate a proper CO tape recorder signal. These results therefore strongly suggest that the origin of the CO tape recorder is mainly derived from variations in tropical CO concentrations in the upper troposphere, which in turn are mainly driven by the seasonality of biomass burning emissions. It is interesting that there are maxima in the CO perturbation over 18 km. These may be mainly caused by transport and chemical effects, rather than just by the seasonal cycle in CO emissions, and deserve further investigation.

[25] For tropospheric O<sub>3</sub> prediction, the contribution of cross-tropopause ozone fluxes (or STE fluxes) to the ozone budget remains relatively uncertain in global CCMs and CTMs [e.g., *Kentarchos and Roelofs*, 2003; *Stevenson et al.*, 2006; *Wild*, 2007]. Whether a CCM has a reasonable cross-tropopause ozone flux is crucial for tropospheric ozone predictions. Table 2 lists the tropospheric O<sub>3</sub> budget terms (i.e., chemical ozone production, chemical ozone loss, surface O<sub>3</sub> deposition, and stratospheric O<sub>3</sub> input fluxes) in runs COUPLED and TROP. The tropospheric ozone budgets are calculated below the (chemical) tropopause, which is defined by the ozone mixing ratio of 150 ppbv. Note that changing the ozone threshold of 150 ppbv to 100 or 200 ppbv has only a small impact on the ozone budget terms. We can see from Table 2 that the stratospheric O<sub>3</sub> input flux in run TROP with tropospheric chemistry only (i.e., STOCHEM) is significantly larger than that in the fully coupled S/T-CCM. The 10 year averaged stratospheric O<sub>3</sub> input flux is 860 Tg yr<sup>-1</sup> in TROP and is 620 Tg yr<sup>-1</sup> in COUPLED. The STE ozone flux cannot be directly observed, but indirect observational constraints indicate a value of 540±140 Tg yr<sup>-1</sup> [*McLinden et al.*, 2000; *Olsen et al.*, 2001]. The ozone STE flux in the fully coupled S/T-CCM is closer to the mean of STE fluxes in the range of CCMs and CTMs [*Stevenson et al.*, 2006], while the STE flux in run TROP is rather high. As the STE flux in run TROP is larger than in run COUPLED, the tropospheric ozone loss in



**Figure 8.** Zonal mean O<sub>3</sub> differences (ppmv) between run TROP and run COUPLED.

run TROP is also larger, while the chemical ozone productions in runs COUPLED and TROP are similar.

[26] As the STE ozone fluxes in run TROP (STOCHEM) and run COUPLED (S/T-CCM) are different, it is necessary here to also compare the tropospheric O<sub>3</sub> simulated by runs COUPLED and TROP. Figure 8 shows O<sub>3</sub> differences between these two runs. We can see that ozone mixing ratios in run TROP are obviously larger than those in run COUPLED. The ozone differences are much larger in the UTLS region with a magnitude of around 1 ppmv, as is also evident in Figures 4d–4f. At the surface, the differences are about 1–5 ppbv. Figure 8 and Table 2 suggest that a 230 Tg yr<sup>-1</sup> increase in O<sub>3</sub> STE flux in the model can cause an increase in lower tropospheric ozone of ~1–5 ppbv. It should be pointed out that the coupling of two chemistry schemes also affects the tropospheric OH distribution and hence the methane lifetime, which is 3% lower in COUPLED compared to TROP (Table 2). This is the opposite to what might be expected from the lower levels of tropospheric O<sub>3</sub> in the COUPLED run and implies that other changes, such as different NO<sub>x</sub> or UV, must be the more dominant control of OH.

## 5. Summary and Conclusions

[27] Combining the UK Met Office Unified Model, the STOCHEM tropospheric chemistry module, and the SLIMCAT stratospheric chemistry module, a chemistry-climate model (S/T-CCM) for the whole atmosphere has been set up and evaluated. The S/T-CCM can be considered as STOCHEM with a much improved, interactive upper boundary or as UMSLIMCAT with a detailed treatment of tropospheric chemistry. The effect of coupling on tropospheric ozone and CO predictions as well as the STE flux of ozone has been discussed through three simulations with different chemistry configurations in the model.

[28] The fully coupled S/T-CCM is computationally efficient and shows good performance in simulating both the stratospheric and tropospheric chemistry compared to various observations. Coupling of two chemistry schemes significantly improves ozone and CO predictions in the UTLS region. The modeled column ozone is in a good agreement with TOMS data, and ozone profiles at different latitudes compare well with UARS and MIPAS climatologies. The ozone and CO correlations are in accordance with the ACE observations in the whole atmosphere, and the observed CO tape recorder signal in the tropical UTLS region is successfully simulated.

[29] The STE ozone flux in the coupled S/T-CCM is close to the mean of the various values in existing global CCMs and CTMs and shows improvements compared with uncoupled STOCHEM. The STE ozone flux in the S/T-CCM has a significant effect on the tropospheric ozone and OH abundance. A higher STE ozone flux in the uncoupled model (TROP) corresponds to higher tropospheric ozone as expected. However, a lower methane lifetime in the coupled model S/T-CCM suggests that changes in UV flux, NO<sub>x</sub>, or other chemical species may have a complex impact on OH in the tropical troposphere. This issue merits further investigation.

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