Future changes in stratosphere-troposphere exchange and their impacts on future tropospheric ozone simulations

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[1] We assess future climate change impacts on stratosphere-troposphere exchange (STE) and their influences on tropospheric O_3, using a chemistry coupled climate model. Tropospheric O_3 distribution and budget were predicted decadally for 1990 to 2100 with emission changes (for O_3 precursors) and climate change specified by the IPCC SRES-A2 scenario. Our simulations show increases in stratospheric O_3 transport to the troposphere as a result of enhancement in the tropospheric and stratospheric circulation with climate change in the model. With emission changes only, net stratospheric O_3 input to the troposphere was simulated to decrease by ~20% during 1990–2100, but to increase by ~80% with including climate change also. Simulated increases in net cross-tropopause O_3 transport are most significant particularly after 2050. *INDEX TERMS:* 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 1620 Global Change: Climate dynamics (3309); 3334 Meteorology and Atmospheric Dynamics: Middle atmosphere dynamics (0341, 0342). **Citation:** Sudo, K., M. Takahashi, and H. Akimoto, Future changes in stratosphere-troposphere exchange and their impacts on future tropospheric ozone simulations, *Geophys. Res. Lett.*, *30*(24), 2256, doi:10.1029/2003GL018526, 2003.

1. Introduction

[2] Many previous studies have suggested that tropospheric ozone (O_3) increases significantly since preindustrial times, in accordance with dramatic increases in anthropogenic emissions especially in the northern hemisphere (NH) [*WMO*, 1990; *Crutzen and Zimmermann*, 1991; *Marenco et al.*, 1994]. Since additional increases in tropospheric O_3 are anticipated in future with probable emission increases in the developing regions like eastern Asia, an accurate prediction of future tropospheric O_3 trend is needed. Future tropospheric O_3 appears to depend largely on emissions of precursor gases (NO_x, CO, and hydrocarbons) and CH_4. However, the effect of future climate change should be also taken into account for future O_3 distributions, since tropospheric O_3 chemistry and transport processes are much affected by meteorological conditions such as water vapor, temperature, clouds, and atmospheric dynamics. Some previous studies suggested that future tropospheric O_3 increases owing to emission changes may be reduced by water vapor increases associated with climate change (warming effect) [e.g., *Fuglestvedt et al.*, 1995; *Stevenson et al.*, 2000; *Johnson et al.*, 2001]. On the other hand, the studies of *Zeng and Pyle* [2003] and *Collins et al.* [2003] have shown increases in downward cross-tropopause O_3 transport with climate change for ~2100 in their simulations, which imply enhanced stratosphere-troposphere exchange (STE) in future. O_3 input from the stratosphere certainly has a large contribution to the O_3 distributions in the tropopause region, despite of its relatively minor contribution to the global tropospheric O_3 source as deduced by a number of modeling studies. Since O_3 changes in the tropopause region cause the largest radiative effect (particularly on surface temperatures) [*Lacis et al.*, 1990], climate change impacts on STE can play a crucial role in future radiative forcing from O_3. In this paper, we investigate the sensitivity of STE to future climate change, simulating global distribution and budget of tropospheric O_3 from 1990 to 2100 considering climate change specified by the IPCC SRES-A2 scenario.

2. Model and Experiments

2.1. The CHASER Model

[3] This study employs the coupled tropospheric chemistry climate model CHASER [*Sudo et al.*, 2002a] which has been developed in the framework of the Center for Climate System Research/National Institute for Environment Studies (CCSR/NIES) atmospheric GCM [*Numaguti*, 1993; *Numaguti et al.*, 1995]. For this study, the horizontal resolution of T42 (2.8° × 2.8°) is adopted with 32 vertical layers from the surface to about 40 km altitude (~1 km vertical resolution in the upper troposphere and lower stratosphere, UTLS). The model considers a detailed on-line simulation of tropospheric chemistry involving O_3-HO_x-NO_x-CH_4-CO system and oxidation of nonmethane hydrocarbons (NMHCs) with a timestep of 10 min, and includes detailed dry and wet deposition schemes also. The CHASER model version adopted in this study is basically identical to that described in *Sudo et al.* [2002a]. However, this version of CHASER, based on the CCSR/NIES GCM version 5.6, newly includes an improved wet deposition scheme, heterogeneous reactions on aerosols for N_2O_5 and several radicals like HO_2, and an on-line simulation of sulfate also. The implemented sulfate simulation is linked to the heterogeneous reactions considered in the model, but not to the GCM's radiation component in this version. In CHASER, advective transport is simulated with a 4th order flux-form advection scheme of the monotonic van Leer [*van Leer*, 1977] and the flux-form semi-Lagrangian scheme of *Lin and Rood* [1996]. Vertical transport associated with

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moist convection is simulated in the framework of the cumulus convection scheme in the AGCM. The model calculates the concentrations of 53 chemical species with 139 reactions (gas/liquid-phase and heterogeneous). The concentrations of stratospheric O_3 and NO_y species above 55 hPa (~20 km) altitude are nudged to the monthly mean satellite data from the Halogen Occultation Experiment project (HALOE) [*Russel et al.*, 1993] and output data from the 3-D stratospheric chemistry model [*Takigawa et al.*, 1999] with a relaxation time of three days. Note that the model does not include the oxygen photolysis and halogen chemistry which would affect lower stratospheric O_3 abundances below 20km altitude in this study. In the detailed model evaluation [*Sudo et al.*, 2002b], good agreements between the CHASER simulations and observations are found for O_3 and precursor species including HO_x radicals.

2.2. Experiments

[4] We set up time-slice simulations for every ten years from 1990 to 2100 with the IPCC SRES A2 scenario [*Houghton et al.*, 2001]. To evaluate impacts of emission change and of climate change separately, this study conducts two experiments: (Exp1) a control experiment only with emission changes and (Exp2) a climate change experiment with emission changes. In Exp1, the GCM simulates present-day meteorological conditions, but in Exp2 it simulates climate change using the A2 scenario. In Exp2, concentrations of CO_2, N_2O, and HFCs for the GCM radiation computation are prescribed by the A2 scenario, while O_3 and CH_4 concentrations are calculated on-line in the chemistry component in the model. Exp2 also includes other forcing factors such as sea surface temperatures (SSTs) and sea ice distributions prescribed by the transient simulations with the CCSR/NIES coupled atmosphere-ocean GCM [*Emori et al.*, 1999; *Nozawa et al.*, 2001] (with no flux adjustment) for the SRES A2 scenario; Exp1 calculates the same meteorological conditions through 1990 to 2100, using the identical SSTs given by the coupled GCM with the present-day climate forcing. Future anthropogenic emissions of O_3 precursors (NO_x, CO, and NMHCs), CH_4, and SO_2 are specified by the A2 scenario. Natural emissions from vegetation, soil, and ocean, considered as in *Sudo et al.* [2002a], are kept constant during 1990–2100. Lightning NO_x emissions, parameterized with the GCM convection, are adjusted to 5 TgN/yr in Exp1, but are prognostically predicted in Exp2. This study also simulates radon distribution following *Sudo et al.* [2002a]. The model was time-integrated for two years including one year spin-up with respect to individual years of 1990, 2000, 2010, . . ., 2100.

3. Results and Discussion

[5] Let us first discuss the changes in net global stratospheric O_3 transport to the troposphere (STE O_3 flux) simulated in Exp1 and Exp2 (Figure 1). Exp1 calculates decreases in net STE of O_3 during 1990–2100 with showing a ~20% decrease in 2100. These reflect increases in tropospheric O_3 export to the stratosphere due to enhanced chemical O_3 production within the troposphere with the emission increases; tropospheric O_3 burden increases by ~50% during 1990 to 2100 in Exp1. In contrast, Exp2 (with climate change) shows increases in O_3 STE calculating a

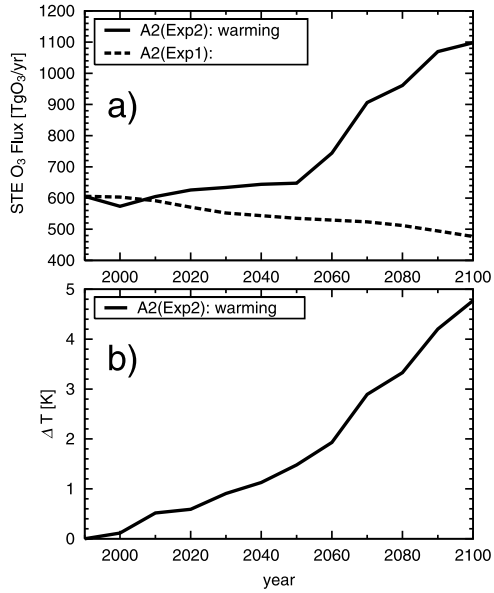

Figure 1. Temporal evolutions of (a) net STE O_3 flux to the troposphere for 1990–2100 calculated in the climate change experiment (Exp2, solid) and in the control experiment (Exp1, dashed) and (b) global and annual mean surface air temperature change in Exp2 relative to 1990. Net STE is shown on a global and annual basis (TgO_3/yr). For the STE estimation, the troposphere is defined to extend from the surface to 100 hPa altitude for |latitude| $\leq 35°$ and to 250 hPa for |latitude| $> 35°$ in the model. The net O_3 STE is assumed to balance with the net O_3 production within the troposphere and surface dry deposition.

STE of as much as 1100 TgO_3/yr in 2100 (+83% relative to 1990). This suggests that future climate change induces increases in downward cross-tropopause O_3 flux overcoming enhancements in upward tropospheric O_3 transport with emission increases. Exp2 calculates more rapid O_3 STE increases after 2050 than before 2050. These coincide with the increases in global mean surface temperature simulated by the GCM in Exp2 (Figure 1b); climate change in the CCSR/NIES GCM with the A2 scenario proceeds rapidly after 2050 due to rapid changes in cloud radiative forcing as reported in *Nozawa et al.* [2001].

[6] The O_3 STE increases simulated in Exp2 can be attributed to the meridional circulation changes induced by the climate change in the model. Figure 2a illustrates the climate change impacts on the residual mean (transformed Eulerian-mean) meridional circulation for 2100 with the temperature increases relative to 1990. The model calculates increases in the stratospheric residual circulation (the Brewer-Dobson circulation) in both hemispheres as in the doubled CO_2 experiment of *Rind et al.* [2001], which enhance the transport from the tropics to the extratropical lower stratosphere. Associated with the residual circulation changes, there is increased ascent in the tropics and increased descent in the subtropical lower stratosphere; e.g., ascent/descent in the tropics/subtropics at 15 km altitude increases by 50–100% (Figure 2b). The model results show large circulation changes also in the troposphere ($z < 15$ km). There is enhanced ascent in the tropics above the altitudes of peak temperature increases (~9 km)

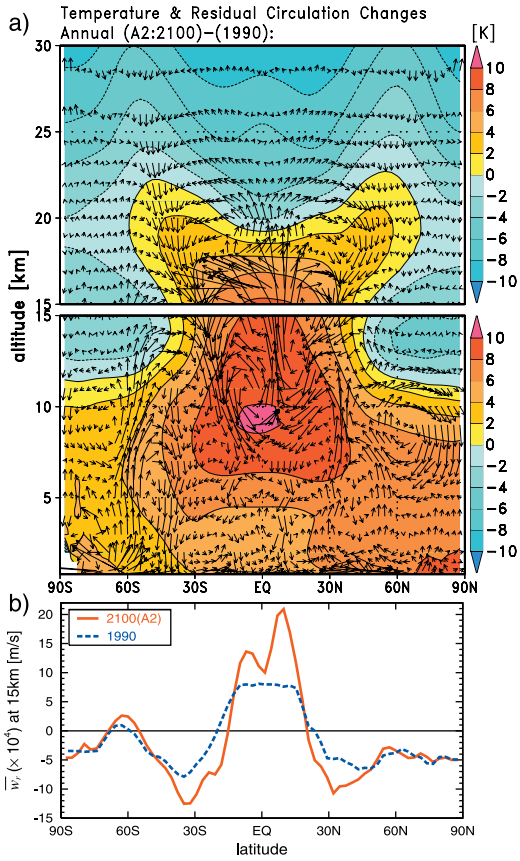

Figure 2. Climate change induced changes in annual and zonal mean temperatures (contours) and in residual mean meridional circulation ($\Delta\overline{v_r}$, $\Delta\overline{w_r}$) (arrows) for 2100 (a); residual mean vertical velocities ($\overline{w_r}$) at 15 km altitude for 2100 (Exp2) and 1990 (b). The changes in (a) are shown as differences between the A2-2100 run of Exp2 (with climate change) and the 1990 run (Exp1). The residual mean meridional circulation ($\overline{v_r}, \overline{w_r}$) is calculated as $\overline{v_r} = \overline{v} - \rho_0^{-1}\left(\rho_0 \overline{v'\theta'}/\overline{\theta}_z\right)_z$, $\overline{w_r} = \overline{w} + (a\cos\phi)^{-1}\left(a\cos\phi \overline{v'\theta'}/\overline{\theta}_z\right)_\phi$ with a the earth's radius, ϕ latitude, θ the potential temperature, and ρ_0 the basic atmospheric density (the subscripts z and ϕ denote vertical and latitudinal derivatives, respectively). Note that different vector (arrow) scales are used for ($\Delta\overline{v_r}$, $\Delta\overline{w_r}$) in the upper (z > 15 km) and lower (z < 15 km) panels of (a) (the vertical component $\Delta\overline{w_r}$ is scaled up by a factor of 1000).

to the lower stratosphere, while enhanced descent is seen around the subtropical jets (latitude ~30°, z ~ 10–15 km). These changes arise from the enhanced Hadley circulation associated with the increases in latitudinal temperature gradient especially in the middle to upper troposphere; as evidence, the zonal wind speeds in the subtropics (subtropical jet) increase by 10–20% in 2100 in Exp2 relative to 1990.

[7] We evaluate the impacts of climate change and associated STE increase on the tropospheric O_3 distribution. Figure 3 gives the differences in O_3 distribution between with and without climate change (Exp2 minus Exp1) with the annual mean O_3 distribution for 2100 in Exp1. As previous studies [e.g., *Johnson et al.*, 2001] suggest, O_3 levels in the lower-middle troposphere are generally reduced resulting from enhanced chemical O_3 destruction due to the increases in water vapor (main cause of O_3 loss) and temperature in Exp2; the net chemical O_3 production within the troposphere estimated for 2100 is 740 TgO_3/yr in Exp1 but −4.7 TgO_3/yr in Exp2. In the upper troposphere, O_3 increases reaching +5–10 ppbv are seen due to the enhanced O_3 input from the stratosphere along with the residual circulation changes as discussed above. O_3 increases in the upper troposphere in NH are small compared with those in SH, reflecting the shorter chemical lifetime of O_3 and larger water vapor increases in NH. In the tropical UTLS, O_3 levels decrease as a result of the stronger ascent (upwelling) associated with the increases in the Hadley and Brewer-Dobson circulations as also seen in the simulations of *Collins et al.* [2003] and *Zeng and Pyle* [2003]. There are O_3 increases of +50–200 ppbv in the extratropical stratosphere originating from the residual circulation changes in the stratosphere (Figure 2). The doubled CO_2 simulation of *Zeng and Pyle* [2003] suggests reduction in O_3 destruction by HO_x in the lower stratosphere due to temperature decreases, which contributes to their calculated increases in lower stratospheric O_3. Our results, however, do not show such an effect, calculating higher O_3 loss rates due to water vapor increases in the lower stratosphere in Exp2 than in Exp1. Our results also show significant O_3 decreases in the lowermost stratosphere (near the tropopause) particularly in the high latitudes in both hemispheres, suggesting increased transport of low O_3 from the lower troposphere. These O_3 decreases appear related to the rises in the tropopause height in Exp2 relative to 1990 (Exp1). Our simulation of atmospheric radon in Exp2 indicates enhanced

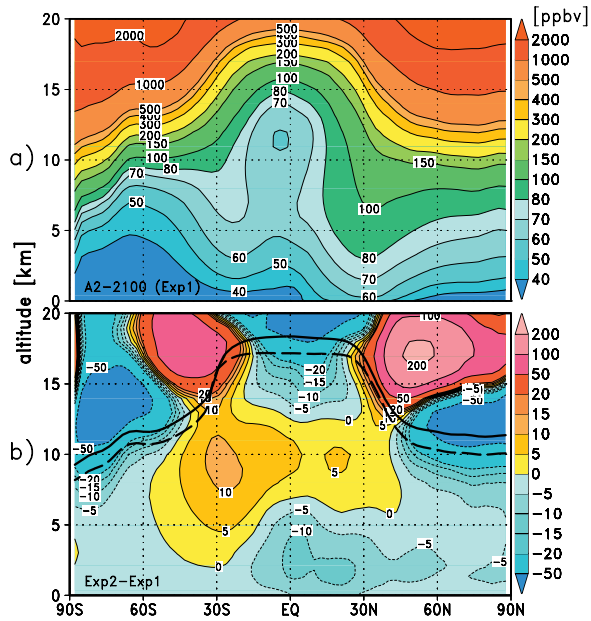

Figure 3. Annual and zonal mean ozone distribution for 2100 calculated in Exp1 (a); climate change induced changes in the ozone distribution (shown as Exp2 minus Exp1) for 2100 (b). In (b), the zonal mean tropopause (determined from the −2 K km^{-1} lapse rate) is also shown for 2100 in Exp2 (solid line) and 1990 in Exp1 (dashed line).

vertical transport from the surface to the UTLS due to the tropospheric circulation changes and increased deep convection associated with the higher tropopause levels (not shown). Similar decreases in lower stratospheric O_3 in the high latitudes are also calculated in *Collins et al.* [2003].

4. Conclusions

[8] We have investigated the impacts of future climate change on stratosphere-troposphere exchange (STE), simulating tropospheric O_3 distributions for 1990 to 2100 with and without climate change using the IPCC SRES A2 scenario. With emission changes only (a control experiment), the net STE O_3 influx to the troposphere decreases by 20% in 2100 responding to the tropospheric O_3 increases due to enhanced chemical O_3 production within the troposphere, while it increases by 83% in 2100 ($\sim$600 in 1990 to 1100 TgO_3/yr in 2100) with climate change as well. Rapid increases in O_3 STE are calculated particularly after 2050 in the climate change experiment (Figure 1), reflecting the climate sensitivity of the CCSR/NIES GCM [*Nozawa et al.*, 2001]. The key factor controlling the STE O_3 increases in the climate change experiment is the change in the residual circulation in the model. We found increases in the Brewer-Dobson circulation and the Hadley circulation due to climate change, which cause enhanced ascent in the tropics and descent in the subtropical lower stratosphere. In response to the increased O_3 STE, upper tropospheric O_3 increases in the low-mid latitudes. The influence of the enhanced O_3 STE on tropospheric O_3 distribution is more significant in SH than in NH, owing to shorter chemical lifetime of O_3 along with larger water vapor increases in NH.

[9] In this study, we have examined the sensitivity of STE to climate change adopting the SRES A2 scenario. It should be noted that the A2 scenario, a high case, predicts relatively large increases in the greenhouse gases and emissions of O_3 precursors. Furthermore, the CCSR/NIES coupled ocean GCM used in this study is considered to have a high climate sensitivity [*Nozawa et al.*, 2001]. In view of these notes, we need further evaluate our climate change simulation. In addition, future changes in stratospheric O_3 distributions, which are not considered in this study, should be included in fact for an accurate prediction of future tropospheric O_3 distributions. For example, future decreases in halogen (mainly chlorine) concentrations are expected to increase lower stratospheric O_3 abundances and hence future O_3 STE trend. On the other hand, halogen-induced O_3 loss may be enhanced by future climate change which causes cooling in the stratosphere. These discussions require the model to be coupled with stratospheric chemistry.

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