Simulation of tropospheric ozone changes during 1997-1998 El Niño: Meteorological impact on tropospheric photochemistry

Kengo Sudo and Masaaki Takahashi

Center for Climate System Research, University of Tokyo, Tokyo, Japan

Abstract. The impact of the 1997-1998 El Niño event on the tropospheric ozone distribution and related processes is assessed by a simulation using a global chemical model. The changes in tropospheric ozone observed during the 1997-1998 El Niño are considered to have been caused by large-scale processes due to the shift in the tropical convection pattern associated with sea surface temperature (SST) changes and by large emissions of ozone precursors due to pronounced biomass burning in the Indonesian region (mainly in Sumatra and Borneo). This simulation study is focused on the effects of meteorological changes caused by the El Niño, aside from the effects of the Indonesian biomass burning. Our simulation results show that about half of the ozone increase observed over the equatorial western Pacific and Indonesia (6-12 Dobson units in tropospheric column ozone) can be explained by the meteorological effects of the El Niño with the remainder attributed to the Indonesian biomass burning, whereas the ozone decrease over the eastern Pacific (4-8 Dobson units) can be attributed to the meteorological effects alone. Moreover, our study suggests that a change in photochemical processes (e.g., water vapor concentration controlling the lifetimes of ozone and other related species, and wet scavenging associated with convective precipitation) also plays an important role in the tropical ozone distribution during the course of the 1997-1998 El Niño event.

1. Introduction

It has been found that the tropical tropospheric ozone (O_3) distribution characterized by a zonal wavenumber 1 pattern [e.g., *Fishman and Larsen*, 1987; *Ziemke et al.*, 1998] fluctuates strongly correlated with the El Niño Southern Oscillation (ENSO) [*Shiotani and Hasebe*, 1994; *Chandra et al.*, 1998; *Ziemke and Chandra*, 1999; *Thompson and Hudson*, 1999]. In El Niño events, changes in sea surface temperature (SST) cause a shift in the convection pattern of the equatorial Pacific, leading to an increase in rainfall and water vapor (H_2O) over the eastern Pacific and a decrease over the western Pacific and Indonesia. During the 1997-1998 ENSO, positive anomalies in tropospheric column ozone (10-20 Dobson units, DU) were observed in the tropical western Pacific and Indonesia by ozone soundings [*Fujiwara et al.*, 1999, 2000] and satellite-based methods [*Chandra et al.*, 1998; *Ziemke and Chandra*, 1999], whereas anomalies observed in the eastern Pacific are negative (4-8

DU) [*Chandra et al.*, 1998], in a phase as appeared during other ENSO events in the past [*Ziemke and Chandra*, 1999]. In Indonesia, the observed positive anomalies (i.e., increase) in tropospheric column ozone during the 1997-1998 ENSO may be primarily attributed to photochemical ozone production associated with the large emissions of ozone precursors by the extensive forest fires in Sumatra and Borneo during August to October 1997 [*Sawa et al.*, 1998; *Fujiwara et al.*, 1999; *Thompson et al.*, 2001]. However, changes in meteorological conditions in Indonesia during this ENSO event (downward motion, suppressed convection, and dryness) can also be significant contributors to the large-scale ozone increase in Indonesia and the western Pacific as suggested by *Chandra et al.* [1998].

In this study, we have evaluated the meteorological impact of the 1997-1998 El Niño on the tropical tropospheric ozone distribution, using a global chemical model. The 1997-1998 El Niño , the strongest ENSO event of the cen-

tury, provided us a good opportunity to study the relationship between tropical meteorological conditions and the tropical ozone distribution through the convection pattern changes. Furthermore, this El Niño event was also a good chance to evaluate the model capability to simulate changes in the ozone distribution and related photochemistry due to natural meteorological changes. *Hauglustaine et al.* [1999] assessed the effect of the 1997 Indonesian fire emissions on the ozone increase in Indonesia during the 1997-1998 ENSO, using a global chemical model. Though their simulation reproduces the ozone increase in the vicinity of the emission source region in Indonesia as observed, the large-scale ozone increase extending outside the Indonesian region and the large-scale ozone decrease in the eastern Pacific are not simulated because their simulation does not account for the meteorological changes of the 1997-1998 El Niño .

2. Global chemical model

In this study, a three-dimensional global chemical model named CHASER (CHemical AGCM for Study of atmospheric Environment and Radiative forcing) is used. This model, developed on the framework of the Center for Climate System Research/National Institute for Environmental Studies (CCSR/NIES) atmospheric general circulation model (AGCM), is aimed at studying the tropospheric photochemistry and its influences on climate. Detailed description and evaluation of CHASER are presented in *Sudo et al.* [2001a, b]. Information on this model can be also obtained via the CHASER web site (http://atmos.ccsr.u-tokyo.ac.jp/~kengo/chaser).

In CHASER, meteorological processes such as transport due to advection, convection, and subgrid-scale mixing are simulated on-line by the dynamical component of the AGCM. The model accounts for emission sources for 10 species including NO_x (43.8 TgN/yr including 5 TgN/yr from lightning), CO (1227 Tg/yr), and nonmethane hydrocarbons (ethane, propane, ethene, propene, acetone, isoprene, terpenes, and a lumped species). Lightning NO_x is calculated in the convection scheme of the AGCM, according to *Price and Rind* [1992]. CHASER considers wet deposition due to convective precipitation and large-scale precipitation as well as dry deposition. In the model, in-cloud and below-cloud scavenging processes are considered separately. The chemical component of the model simulates the O_3-HO_x-NO_x-CH_4-CO photochemical system and oxidation of nonmethane hydrocarbons through 88 chemical and 25 photolytic reactions with 47 chemical species in its present configuration.

For this simulation, we adopted a horizontal resolution of T21 (5.6° longitude × 5.6° latitude), and a relatively high vertical resolution with 32 layers from the surface up to about 40 km altitude. The model uses a σ coordinate system in the vertical and has a vertical resolution of 1 km in the free troposphere and much of the lower stratosphere.

An evaluation of the model suggests that it is able to simulate the observed tropospheric ozone distribution [*Sudo et al.*, 2001b]. The model reproduces the zonal wavenumber 1 structure in tropical tropospheric column ozone, calculating an ozone peak in the Atlantic (40-48 DU) and a minimum in the western Pacific (15-20 DU) in October as shown in Figure 1. In a climatological simulation by the model, the photochemical production and destruction of O_3 in the global troposphere are estimated at 4895 TgO_3/yr and 4498 TgO_3/yr, respectively (leading to net O_3 production of 397 TgO_3/yr). The stratospheric influx of O_3 is estimated at 593 TgO_3/yr, in the middle of the range of previous studies (391 TgO_3/yr [*Hauglustaine et al.*, 1998] to 846 TgO_3/yr [*Berntsen and Isaksen*, 1997]).

3. Simulation method

In this study, our simulation results are evaluated, using tropical anomalies (October 1997 minus October 1996) in tropospheric column ozone (TCO) derived by a satellite-based method (convective-cloud differential, CCD technique) [*Chandra et al.*, 1998]. We have set up a simulation for 2 years, 1996 and 1997, for a direct comparison with *Chandra et al.* [1998]. To reproduce the meteorological conditions in 1996 and 1997, we used analyzed data of wind velocities and temperature from the European Center for Medium-Range Weather Forecasts (ECMWF) as a constraint to the dynamical component of the model, in addition to sea surface temperature (SST) data for 1996 and 1997. The model is nudged by using the daily ECMWF data with a relaxation time of 1 day. The nudged model generally well reproduces the tropical water vapor distributions in 1996 and 1997 from the ECMWF observations. To evaluate the meteorological impact of the El Niño only, the effect of the 1997 Indonesian fires is not taken into account in this simulation, and the same surface emission rates in Indonesia are used for 1996 and 1997 in the model.

4. Results and discussion

4.1. Impact on tropospheric ozone

Figure 2 shows a comparison between observed and simulated anomalies (October 1997 minus October 1996) in TCO (DU). The observed anomalies are derived from Earth Probe (EP) Total Ozone Mapping Spectrometer (TOMS) using the convective-cloud differential (CCD) technique [*Chandra et al.*, 1998]. TCO calculated by the model shows the

O_3 column integrated from the surface to the tropopause defined as the lowest altitude at which the vertical temperature gradient is greater than -2 K/km in the model. The simulated TCO anomalies in October show an asymmetrical dipole structure centered around the date line with positive anomalies in the western Pacific and Indonesia and negative anomalies in the eastern Pacific, well reproducing the observed changes in TCO in October 1997. The simulated negative anomaly (decrease) in TCO ranges from 2 DU to 8 DU through much of the eastern Pacific, in good agreement with the observed values. The simulation also shows positive TCO anomalies (increase) of 10-12 DU in Indonesia and the Indian Ocean. The peak values of the observed O_3 increase (16-20 DU) are higher than the simulated values (10-12 DU) by 6-8 DU. This underestimation of the simulation may be attributed to the effect of the Indonesian fire emissions [*Fujiwara et al.*, 1999; *Hauglustaine et al.*, 1999; *Thompson et al.*, 2001]. However, it is interesting to note that both the observations and the simulation show a peak over the eastern Indian Ocean and the edge of Sumatra, demonstrating the considerable importance of the El Niño induced meteorological changes for the observed O_3 increase over this region. The observations also show positive anomalies (2-4 DU) over South America, probably reflecting the effect of changes in the zonal circulation (Walker circulation). Although these observed positive anomalies in South America are not clearly seen in the model results for October, similar positive anomalies (4-8 DU) are calculated by the model over South America in November (not shown).

4.2. Implications from simulation

Figure 3(a) shows the simulated changes (October 1997 minus October 1996) in the vertical distribution of O_3 averaged over 10°S-10°N. As expected from Figure 2, an asymmetrical dipole structure is seen over 60°E-100°W, strongly anti-correlated with the changes in vertical wind velocity and convective cloud mass flux computed in the model with the Arakawa-Schubert convection scheme (Figure 3(b)). 70-90% of the simulated TCO changes (Figure 2, bottom) comes from O_3 changes below 10 km as shown in Figure 3(a). The important factors controlling the O_3 increase over the Indonesian region (70°E-170°E) are downward motion, suppressed convection, and dryness as suggested by *Chandra et al.* [1998]. Downward motion, together with suppressed convection, brings O_3 produced in the upper troposphere down, causing O_3 in the lower-middle troposphere to increase. Additionally, the dryness, leading to a longer lifetime for O_3, seems to contribute to the simulated O_3 increase over this region. Since H_2O is the main cause of O_3 loss in the low to middle troposphere [e.g., *Kley et al.*, 1996], the simulated chemical lifetime of ozone increases

by 60-100% in the middle troposphere over 70°E-170°E as shown in Figure 3(d), anti-correlated with the specific humidity changes of Figure 3(c). In the Indonesian region, the chemical lifetime of ozone at 5 km altitude is prolonged typically from $\sim$10 days in October 1996 to $\sim$25 days in October 1997 in the model. The O_3 increase ($>$20 ppbv) in the upper troposphere (higher than 12 km) over the Indonesian region appears to be also associated with the downward motion and suppressed convection leading to less efficient vertical transport of low O_3 air masses from the surface, and possibly to subsidence of stratospheric O_3. These processes are reversed where there is upward motion, enhanced convection, and H_2O increase (170°E-90°W). Upward motion and enhanced convection advect low O_3 and high H_2O upward, resulting in lower O_3 (-5 to -15 ppbv) over this region (Figure 3(a)).

Moreover, the simulation indicates that the photochemical O_3 production process is also affected by the El Niño. Here, we focus our attention on the peak region of ozone increase over 10°S-0°. Figure 4(a) and (b) show longitude-altitude cross sections (10°S-0°) of the net chemical production (P-L) of O_3 (ppbv/day) simulated for October 1996 and 1997, respectively. In 1997, the O_3-producing area (P-L$>$0) is enlarged downward to 3-4 km over Indonesia. The net O_3 production at $\sim$5 km over Indonesia increases by 1-1.5 ppbv/day, changing from negative (sink) in 1996 to positive (source) in 1997. This enhancement in the net O_3 production in the middle troposphere is due to the H_2O decrease as discussed above and also to increases in O_3 precursors such as nitrogen oxides NO_x ($NO+NO_2$) and carbon monoxide CO (Figure 4(c) and (d)) in the low to middle troposphere. The NO_x changes below 8 km appear to be caused by changes in the efficiency of wet deposition of HNO_3 associated with precipitation pattern changes, and also by changes in the NO_x removal by OH (hydroxyl radical) due to H_2O changes. The HNO_3 lifetime against wet deposition averaged over the Indonesian region is prolonged from $\sim$1 day in October 1996 to 3-5 days in 1997, leading to an increase in HNO_3 by 200-500%. NO_x increases in the upper troposphere (above 12 km) over 70°E-170°E may be due to subsidence of stratospheric NO_x. A slight NO_x decrease (5-10 pptv) at 6-12 km over $\sim$110°E may be attributed to a decrease in lightning NO_x production over Indonesia due to suppressed convection in the model. In the case of CO, the simulated increases ($>$10 ppbv) in the lower troposphere over 70°E-170°E appear to be due to suppressed convection and downward motion (Figure 3(b)). Figure 4(d) also shows CO increases in the middle troposphere over 60°E-120°E. These CO increases appear to be associated with a longer lifetime for CO due to reduced OH levels, and also with enhanced transport from South Africa owing to the changes in the circulation

pattern in the tropics as illustrated in Figure 3(b). It should also be noted that the net O_3 production in the upper troposphere (>12 km) over Indonesia is weaken in October 1997 by 20-40% (Figure 4(b)) in spite of the NO_x increase in this region (c). This arises from decreases in other O_3 precursors such as HO_x (OH+HO_2), CO, and hydrocarbons in the model as shown in Figure 4(d). In October 1997, upward transport of CO, hydrocarbons, and HO_x precursors such as formaldehyde, acetone, and peroxides to the upper troposphere over the Indonesian region is not efficient due to suppressed convection and downward motion in this region. In the model, HO_x decreases by 50-70% in the Indonesian upper troposphere in October 1997.

5. Summary and conclusions

In this study, the meteorological impact of the 1997-1998 El Niño on the tropical tropospheric ozone distribution has been investigated using a global chemical model. To evaluate the El Niño induced meteorological impact alone, the effect of the 1997 Indonesian fires is ignored intentionally in this simulation. The simulation generally well reproduces the observed ozone anomalies (increase and decrease) in October 1997, though the peak values in the ozone anomaly over the Indonesian region are underestimated by the model due to lack of consideration of the 1997 Indonesian fire emissions. The key factors controlling the simulated ozone changes are (1) the downward/upward motions, (2) the suppressed/enhanced convection, and (3) the associated water vapor (H_2O) changes in the tropics.

In the simulation, 70-90% of the calculated change in tropospheric column ozone is caused by O_3 changes below 10 km altitude. The simulation shows that the O_3 increases below 10 km over the Indonesian region are related also to enhanced O_3 production in the middle troposphere due to increase in NO_x and other O_3 precursors associated with meteorological changes such as suppressed convection, downward motion, dryness, and reduced precipitation in this region. As *Chandra et al.* [1998] suggests, O_3-producing air mass appears to increase in this region in the model. On the contrary, in the upper troposphere over Indonesia, simulated O_3 production decreases by 20-40% owing to reduced injection of O_3 precursors associated with suppressed convection and downward motion in the region. Consequently this model simulation suggests that the effect of the 1997 Indonesian fire (not considered in this study) on O_3 production is most significant in the lower troposphere (especially in the boundary layer), and is less significant in the upper troposphere.

References

Berntsen, T. K., and I. S. A. Isaksen, A global three-dimensional chemical transport model for the troposphere, *J. Geophys. Res.*, *102*, 21,239–21,280, 1997.

Chandra, S., J. R. Ziemke, W. Min, and W. G. Read, Effects of 1997-1998 El niño on tropospheric ozone and water vapor, *Geophys. Res. Lett.*, *25*, 3867–3870, 1998.

Fishman, J., and J. C. Larsen, Distribution of total ozone and stratospheric ozone in the tropics: Implications for the distribution of tropospheric ozone, *J. Geophys. Res.*, *92*, 6627–6634, 1987.

Fujiwara, M., K. Kita, S. Kawakami, T. Ogawa, N. Komala, S. Saraspriya, and A. Suripto, Tropospheric ozone enhancements during the indonesian forest fire events in 1994 and 1997 as revealed by ground-based observations, *Geophys. Res. Lett.*, *26*, 2417–2420, 1999.

Fujiwara, M., K. Kita, T. Ogawa, S. Kawakami, T. Sano, N. Komala, S. Saraspriya, and A. Suripto, Seasonal variation of tropospheric ozone in indonesia revealed by 5-year ground-based observations, *J. Geophys. Res.*, *105*, 1879–1888, 2000.

Hauglustaine, D. A., G. P. Brasseur, S. Walters, P. J. Rasch, J.-F. Müller, L. K. Emmons, and M. A. Carroll, MOZART, a global chemical transport model for ozone and related chemical tracers, 2., Model results and evaluation, *J. Geophys. Res.*, *103*, 28,291–28,335, 1998.

Hauglustaine, D. A., G. P. Brasseur, and J. S. Levine, A sensitivity simulation of tropospheric ozone changes due to the 1997 Indonesian fire emissions, *Geophys. Res. Lett.*, *26*, 3305–3308, 1999.

Kley, D., P. J. Crutzen, H. G. J. Smit, H. Vömel, S. J. Oltmans, H. Grassl, and V. Ramanathan, Observations of near-zero ozone concentrations over the convective Pacific: Effects on air chemistry, *Science*, *274*, 230–233, 1996.

Price, C., and D. Rind, A simple lightning parameterization for calculating global lightning distributions, *J. Geophys. Res.*, *97*, 9919–9933, 1992.

Sawa, Y., H. Matsueda, Y. Tsutsumi, J. Jensen, H. Y. Inoue, and Y. Makino, Tropospheric carbon monoxide and hydrogen measurements over Kalimantan in Indonesia and northern Australia during October 1997, *Geophys. Res. Lett.*, *26*, 1389–1392, 1998.

Shiotani, M., and F. Hasebe, Stratospheric ozone variations in the equatorial region as seen in Stratospheric Aerosol and Gas Experiment data, *J. Geophys. Res.*, *99*, 14,575–14,584, 1994.

Sudo, K., M. Takahashi, and H. Akimoto, CHASER: A global chemical model of the troposphere 1. Model description, *J. Geophys. Res.*, submitted, 2001a.

Sudo, K., M. Takahashi, and H. Akimoto, CHASER: A global chemical model of the troposphere 2. Model results and evaluation, *J. Geophys. Res.*, submitted, 2001b.

Thompson, A. M., and R. D. Hudson, Tropical tropospheric ozone (TTO) maps from Nimbus 7 and Earth Probe TOMS by the

modified-residual method: Evaluation with sondes, ENSO signals, and trends from Atlantic regional time series, *J. Geophys. Res.*, *104*, 26,961–26,975, 1999.

Thompson, A. M., J. C. Witte, R. D. Hudson, H. Guo, J. R. Herman, and M. Fujiwara, Tropical tropospheric ozone and biomass burning, *Science*, *291*, 2128–2132, 2001.

Ziemke, J. R., and S. Chandra, Seasonal and interannual variabilities in tropical tropospheric ozone, *J. Geophys. Res.*, *104*, 21,425–21,442, 1999.

Ziemke, J. R., S. Chandra, and P. K. Bhartia, Two new methods for deriving tropospheric column ozone from TOMS measurements: Assimilated UARS MLS/HALOE and convective-cloud differential techniques, *J. Geophys. Res.*, *103*, 22,115–22,127, 1998.

K. Sudo and M. Takahashi, Center for Climate System Research, University of Tokyo, 4-6-1, Komaba, Meguro-ku, Tokyo, 153-8904, Japan. (kengo@ccsr.u-tokyo.ac.jp; masaaki@ccsr.u-tokyo.ac.jp)
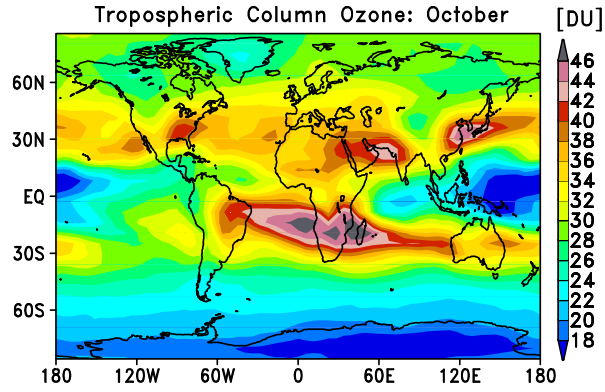
Figure 1. Tropospheric column ozone (TCO) simulated by CHASER for October (Dobson Units, DU).
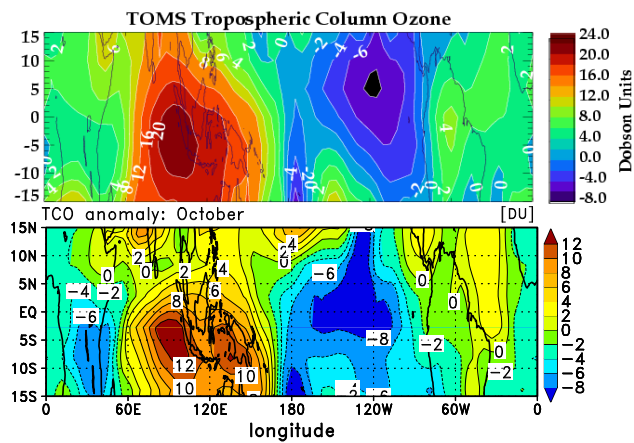

Figure 2. Observed and calculated tropical anomalies (1997 minus 1996) in TCO showing the effects of El Niño (1997). (Top) EP TOMS CCD TCO for October (DU) [*Chandra et al.*, 1998], (bottom) model results for October. The model results show TCO anomalies due to meteorological effects only.
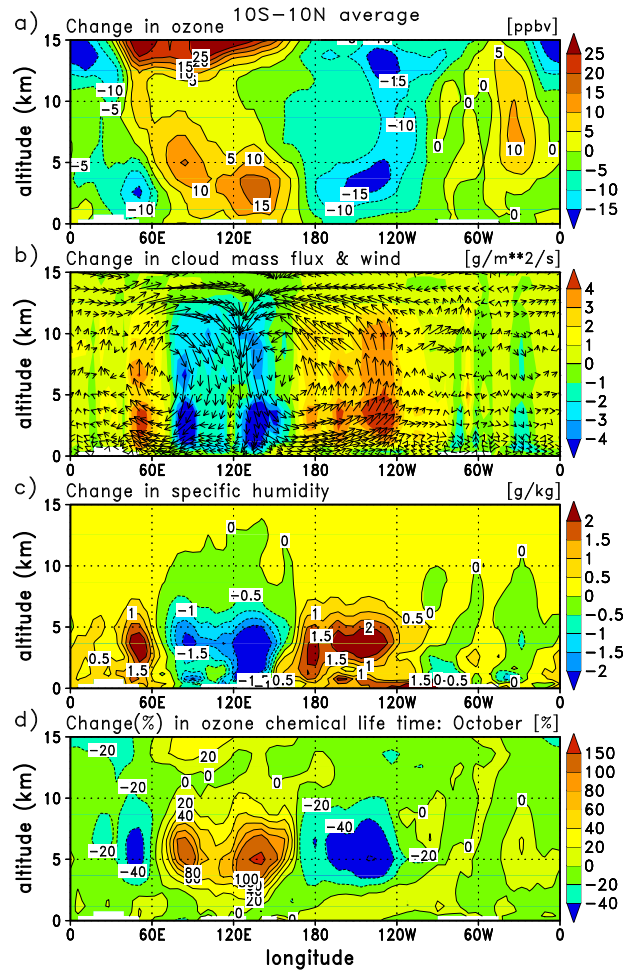
Figure 3. Longitude-altitude cross section of simulated anomalies in (a) ozone (ppbv), (b) convective cloud mass flux (g m^{-2} s^{-1}) and wind vector of zonal and vertical velocity (vertical velocity is scaled up by a factor of 1000), (c) specific humidity (g kg^{-1}), and (d) chemical lifetime of ozone (%) averaged over 10°S-10°N. The differences are shown as October 1997 minus October 1996.
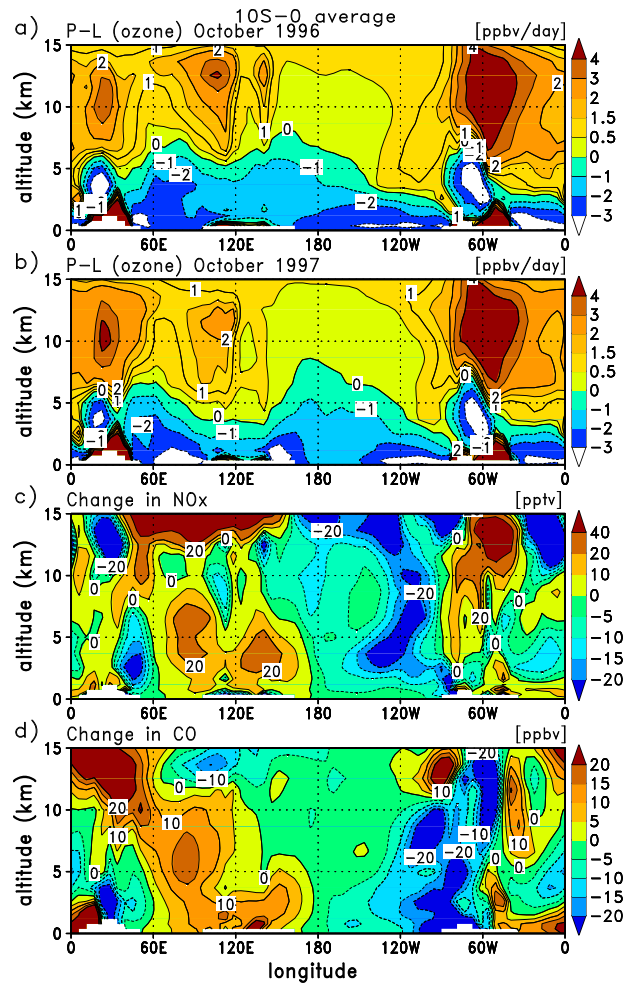
Figure 4. Longitude-altitude cross section (10°S-0°) of net chemical ozone production (ppbv/day) simulated (a) for October 1996 and (b) for October 1997, and simulated anomalies in (c) NO$_x$, and in (d) CO. The anomalies are shown as October 1997 minus October 1996.