CO_2 in the upper troposphere: Influence of stratosphere-troposphere exchange

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[1] A two-dimensional transport model constrained to measured surface CO_2 concentrations was used to simulate the spatial and temporal variation of CO_2 in the atmosphere for the period from 1975 to 2004. We find that the amplitude, phase and shape of the CO_2 seasonal cycle vary as a function of both altitude and latitude. Cross tropopause exchanges, especially the downward branch of the Brewer-Dobson circulation, which brings stratospheric air to the upper troposphere at middle and high latitudes, change the CO_2 concentration and seasonal cycle in the extra-tropics. The model results match recent aircraft measurements of CO_2 in the upper troposphere (*Matsueda et al.*, 2002) remarkably well. We conclude that upper tropospheric CO_2 volume mixing ratios will provide a valuable tool for validating vertical transport. The implications of the CO_2 variation caused by the stratosphere-troposphere exchange for remote sensing of CO_2 are discussed. **Citation:** Shia, R.-L., M.-C. Liang, C. E. Miller, and Y. L. Yung (2006), CO_2 in the upper troposphere: Influence of stratosphere-troposphere exchange, *Geophys. Res. Lett.*, *33*, L14814, doi:10.1029/2006GL026141.

1. Introduction

[2] Currently, the information about the distribution and variation of atmospheric CO_2 and the inferred CO_2 sources and sinks are derived from model simulations and inversions constrained by *in situ* measurements of CO_2, mostly in the lower troposphere [*Fan et al.*, 1998; *Tans et al.*, 1990; *Anderson et al.*, 1996; *Nakazawa et al.*, 1997; *Zahn et al.*, 1999; *Randerson et al.*, 2002; *Law et al.*, 2003; *Gurney et al.*, 2004]. The *in situ* measurements have high accuracy but limited spatial coverage. Satellite measurements provide much better spatial coverage but lower measurement precision [*Buchwitz et al.*, 2005a, 2005b]. Next generation satellites designed specifically to measure atmospheric CO_2 will return column averaged CO_2 mixing ratios under clear conditions with precision of 1 ppmv, e.g., the Orbiting Carbon Observatory (OCO) [*Crisp et al.*, 2004]. CO_2 mixing ratios in the upper troposphere are currently retrieved from the Infrared Atmospheric Sounder (AIRS) data with a precision of 1.2 ppmv [*Chahine et al.*, 2005].

[3] The difference between the column-averaged CO_2 mixing ratio and the surface value varies from 2 to 10 ppmv depending on location and time of year [*Olsen and Randerson*, 2004]. The upper troposphere can contribute significantly to this difference because this portion of the column constitutes approximately 20% of the column air mass and the CO_2 mixing ratio in this region can differ by 5 ppmv or more from the CO_2 mixing ratio at the surface [*Anderson et al.*, 1996; *Matsueda et al.*, 2002]. Since CO_2 is inert in the atmosphere, its trend and seasonal cycle propagate primarily from the surface. The difference of CO_2 mixing ratios between the surface and a given altitude is determined by the processes that transport surface air throughout the atmosphere, including advection, convection and eddy mixing.

[4] In the extra-tropics of the upper troposphere during winter, the CO_2 concentration is particularly influenced by the air descending from the lower stratosphere in the downward branch of the Brewer-Dobson circulation. The CO_2 mixing ratio is lower and the seasonal cycle is different in the stratosphere because it takes more than 1 year to transport surface air to the stratosphere through the tropical pipe [*Plumb and Ko*, 1992; *Plumb*, 1996] and the original seasonal cycle from the surface is diluted [*Mote et al.*, 1996]. Earlier studies of nitrous oxide demonstrate the influence of the stratosphere on its seasonal cycle in the troposphere [*Liao et al.*, 2004; *Nevison et al.*, 2004], caused by the same mechanism discussed above. The stratospheric influence has been detected down to the surface [*Nevison et al.*, 2004; X. Jiang et al., The seasonal cycle of N2O: Analysis of data, submitted to *Global Biogeochemical Cycles,* 2006].

[5] Recently, *Matsueda et al.* [2002] reported measurements of the spatial and temporal variations of atmospheric CO_2 mixing ratios at 8–13 km from samples collected biweekly from a commercial airliner flying between Sydney (Australia) and Tokyo (Japan) area. These data are suitable for testing model transport of air from the surface into the upper troposphere as well as the incursion of stratospheric air back into the upper troposphere. These processes result in the differences between surface and upper tropospheric CO_2 mixing ratios described above.

2. Data and Model Description

[6] We used the Caltech/JPL two-dimensional chemistry transport model (2-D CTM) to simulate CO_2 distributions in the atmosphere. A detailed description of the 2-D CTM is given by *Morgan et al.* [2004]. An important feature of our 2-D CTM is its ability to reproduce the age of air in the stratosphere derived from measurements of CO_2 and SF_6 [*Boering et al.*, 1996; *Morgan et al.*, 2004]. The model uses logarithmic pressure as the vertical coordinate. It has an 18 × 40 latitude-pressure grid extending from pole to pole and from the surface to about 80 km. The stream function of the atmospheric circulation and the horizontal and vertical diffusivities are taken from *Morgan et al.* [2004]. Because CO_2 is non-reactive in the troposphere and lower strato-

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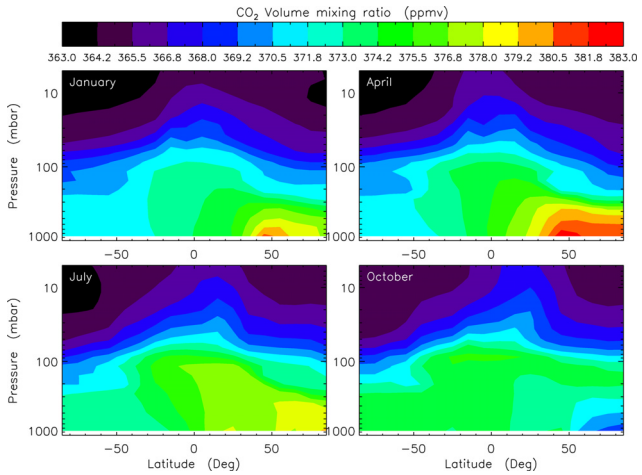

Figure 1. Latitude-pressure plots of modeled CO_2 mixing ratios for January, April, July and October of 2003.

sphere while photolysis above 50 km negligibly affects the total stratospheric CO_2 mixing ratio, the model includes no CO_2 chemistry.

[7] The observed surface CO_2 mixing ratios from 1975 to 2004 have been used as the lower boundary condition for the 2-D CTM. We assume that all atmospheric CO_2 originates from the surface layer and constrain the model using CO_2 mixing ratios from a complete and accurate proxy record of CO_2 of the National Oceanic and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL) [*Tans et al.*, 1998], which has compiled the surface CO_2 mixing ratio since 1975 at 76 latitudes (55 static platforms plus 21 ship measurements) from the South Pole to 82.45°N (Alert, Nunavut, Canada). The data are gridded uniformly in latitude to fit into our model resolution. We reconstruct missing data from before 1990 by taking either the data in the same month from previous and subsequent years (for $\sim$1 year missing data) or the record from the closest available sites (for $\gg$1 year missing data). There is no significant missing data after 1990. The lower boundary condition of the model is updated each monthly time step. This time resolution is sufficient for studying the seasonal cycle and longer-term variations.

[8] Comparisons of the 2-D CTM results with the *Matsueda et al.* [2002] data were made to validate the model. The aircraft experiments sampled air at latitudes from 30°S to 30°N (Sydney to Tokyo) and altitudes from 8 to 13 km from April 1993 to March 2005.

3. Model Results and Discussion

[9] The observed surface CO_2 mixing ratios from 1975 to 2004 have been used as the lower boundary condition for the 2-D CTM. The observed surface concentrations act as a net source of CO_2 in the atmosphere. The model then propagates surface air into the free troposphere and the stratosphere. The modeled CO_2 mixing ratios in January, April, July and October of 2003 are plotted as a function of latitude and pressure in Figure 1. The CO_2 mixing ratio at all altitudes in the tropical troposphere is close to that at the surface due to large vertical and horizontal diffusion coefficients, which are used in the model to parameterize the fast convective and eddy mixing in the tropics. In the extratropics, the CO_2 mixing ratio is affected by the cross tropopause exchange and thus has a larger vertical gradient. The effect of cross tropopause transport caused by the downward branch of the Brewer-Dobson circulation on the CO_2 mixing ratio is clearly seen at high latitudes.

[10] Figure 2 compares the CO_2 mixing ratios between 9 km and 13 km from the model simulation and aircraft observations for the period from 1994 to 2004. The two layers at 273 mb and 205 mb encompass the region of 9 km to 13 km in the low latitudes in our model and we take the average of CO_2 mixing ratio in the two layers to represent this region. The aircraft data have been re-sampled to match the meridional grid of our 2-D CTM. The model reproduces the observations very well, especially for 15°N, 25°N, and 35°N. The corresponding CO_2 mixing ratios at the surface [*Tans et al.*, 1998] are also plotted for comparison. Differences of 2 to 6 ppmv between the surface and the upper tropospheric CO_2 mixing ratios can be seen. Also their seasonal cycles are different in amplitude, phase and shape. The change of the seasonal cycle reflects the seasonality of the cross tropopause exchange, which is larger during winter and spring. This feature can be seen clearly in Figure 2, especially at higher latitudes of the Northern Hemisphere. In the Southern Hemisphere, there is little difference in CO_2 mixing ratios between the surface and the upper troposphere because of the weakness of the Brewer-Dobson circulation [*Appenzeller et al.*, 1996].

[11] We performed two sensitivity tests to understand the role of the Brewer-Dobson circulation, which is the main process for stratosphere-troposphere exchange. We increased the advection by 50% in one test and decreased it

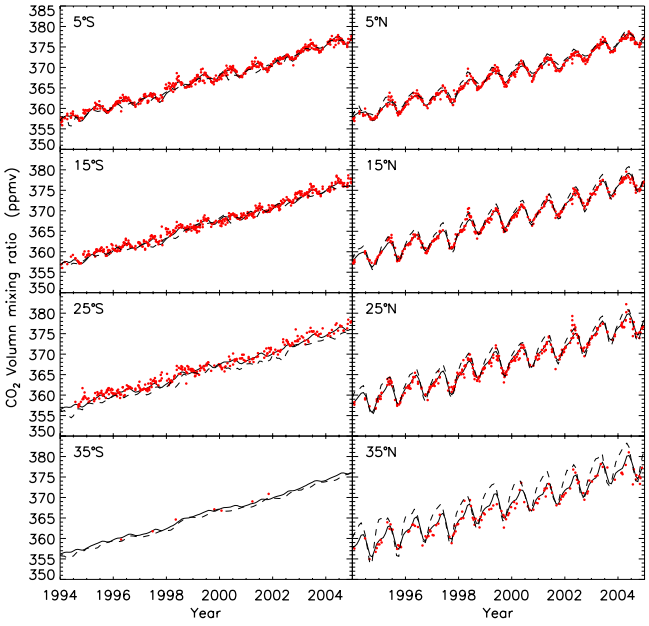

Figure 2. Aircraft observations between 9 km and 13 km (red dots) [*Matsueda et al.*, 2002] and modeled CO_2 mixing ratios averaged at the layer between 9 km and 13 km (solid line) from 1994 to 2004. The panels are for 35°S, 25°S, 15°S, 5°S, 5°N, 15°N, 25°N, and 35°N, respectively. For comparison, the surface CO_2 mixing ratios [*Tans et al.*, 1998] are shown by dashed lines.

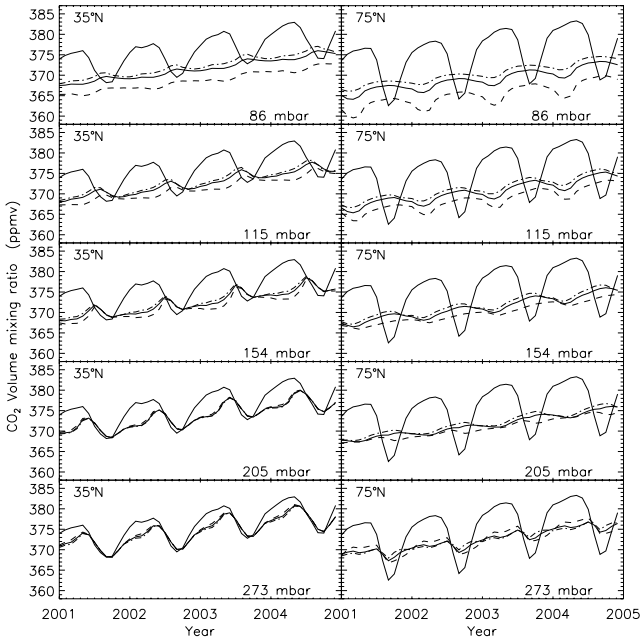

Figure 3. Modeled CO_2 mixing ratios for the standard case (solid lines), test run A (using 50% reduction in advection, dashed lines), and test run B (using 50% enhancement in advection, dash-dotted lines) at the 86, 115, 154, 205, and 273 mb levels. (left) For 35°N and (right) for 75°N. The surface CO_2 mixing ratio is also plotted for comparison (triple-dot-dashed lines).

by 50% in the other. The results from the two test cases are plotted along with the baseline simulation (solid lines) in Figure 3 for 35°N and 75°N. In the model the tropopause at 35°N is roughly located between the 115 mb layer and the 154 mb layer. At 75°N. it is between the 205 mb layer and the 273 mb layer. Let us first examine the results for 75°N at 86 mb, 115 mb and 154 mb, where the main effect is the age of air. As we increase the Brewer-Dobson circulation, it takes less time for the air to reach the stratosphere. The age of air in this case is younger than that in the standard case. Therefore, the mixing ratio of CO_2 (dash-dotted lines) is higher than that for the standard case (solid lines). The reverse happens when the Brewer-Dobson circulation is reduced, as indicated by the dashed lines. For comparison, the CO_2 mixing ratio at the surface is also plotted (triple-dot-dashed lines) in each panel.

[12] Lower down in the atmosphere (Figure 3, 273 mb at 75°N), the overall impact of the Brewer-Dobson circulation is less significant due to the dilution of stratospheric air by tropospheric air. The seasonal variability of CO_2 becomes more important. In the stratosphere the Brewer-Dobson circulation attains maximum strength in later winter and early spring [*Appenzeller et al.*, 1996], resulting in a CO_2 minimum in spring (e.g., see solid line in Figure 3, 86 mb at 75°N). This spring minimum propagates from the stratosphere to the troposphere with a phase lag. However, at the surface the seasonal cycle of CO_2 is regulated by the biosphere [*Tans et al.*, 1998]. A minimum in CO_2 mixing ratio is attained in late summer. As the influence of the Brewer-Dobson circulation is reduced, most of the change appears as a shift in the seasonal cycle of CO_2. This is apparent in Figure 3, 75°N at 273 mb (dashed and dash-dotted lines).

[13] At lower latitude Figure 3 (35°N) shows that the model results are qualitatively similar to those for 75°N, but the amplitudes for the stratospheric effects are reduced.

[14] In Figure 4, column averaged CO_2 mixing ratios calculated for the year 2003 are plotted in the left column for 45°S, 15°N, 35°N, and 85°N with the surface to CO_2 mixing ratios to compare their seasonal cycles. The amplitude of the seasonal cycle in the column averaged CO_2 mixing ratio is smaller than that in the surface CO_2 mixing ratio. This difference has been discovered previously by studies using a 3-D model [e.g., see *Olsen and Randerson*, 2004, Figure 6]. However, the shape of the seasonal cycle simulated by our model does not follow the surface cycle as closely as the 3-D model does. We believe this is due to the influence of the stratosphere in our 2-D CTM. In Figure 4 (right), the difference between the 2-D model computed column averaged CO_2 mixing ratios and the surface CO_2 mixing ratios at 45°S, 15°N, 35°N, and 85°N for the same year is plotted. The variation in this difference at middle and high latitudes reflects the seasonality of troposphere-stratosphere exchange. The difference is about 0.5 ppmv at 15°N, 3 ppmv at 35°N, and 7 ppmv at 85°N. The contribution to this difference from the layer between 9 and 15 km is also plotted in Figure 4 (right). It can be seen that the upper troposphere makes little contribution to this difference at 45°S and 15°N, but contributes 1 ppmv at 35°N, and 2 ppmv at 85°N. The OCO mission is designed to retrieve column CO_2 values with 1 ppmv precision and should be able to detect these signals, at latitudes north of 35°N.

4. Conclusions

[15] Our study shows that the extra-tropical CO_2 mixing ratios in the upper troposphere and at the surface differ

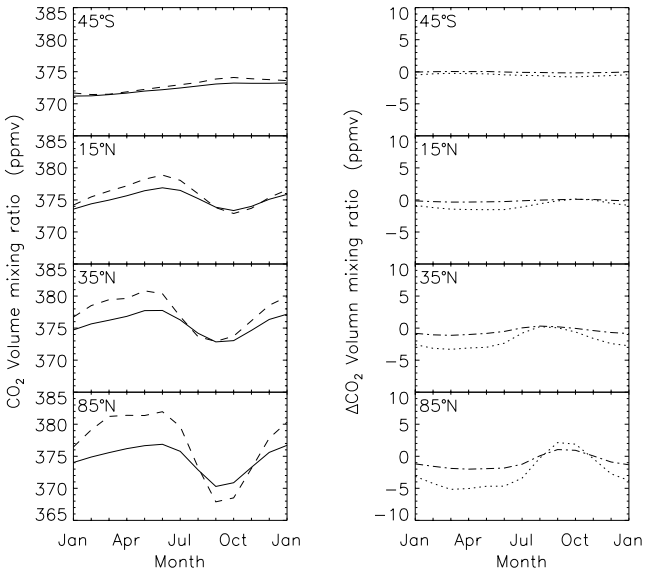

Figure 4. (Left) The model column averaged CO_2 mixing ratios (solid lines) and the surface CO_2 mixing ratios (dashed lines) at 45°S, 15°N, 35°N, and 85°N from top to bottom. (Right) The difference between the model calculated column averaged CO_2 mixing ratios and the surface CO_2 mixing ratios (dotted lines), and the contribution of the layer between 9 and 15 km to this difference (dash-dotted lines) at 45°S, 15°N, 35°N, and 85°N from top to bottom.

significantly in the amplitude, phase and shape of the seasonal cycle. Modeled CO_2 mixing ratios in the upper troposphere are in excellent agreement with *in situ* aircraft observations and are also consistent with the CO_2 retrievals from (The Television and Infrared Operational Satellite-Next Generation (TIROS-N) Operational Vertical Sounder [*Chédin et al.*, 2003]. The averaging kernel of the Atmospheric Infrared Sounder has maximum sensitivity to CO_2 in the 8–13 km range [*Chahine et al.*, 2005] and should provide excellent global data for comparison with our predicted variations.

[16] The comparison of our model results with aircraft observations demonstrates that 2-D models can accurately simulate altitude and latitude variations in atmospheric CO_2 mixing ratios. Similar studies employing 3-D models should provide additional insight into the global carbon cycle and are essential for inferring the location of CO_2 sources and skinks. However, some of the current 3-D model studies underestimates the amplitude of the seasonal cycle in the column CO_2 mixing ratio [*Olsen and Randerson*, 2004; *Warneke et al.*, 2005]. Our study suggests that part of this discrepancy may result from the model's lack of realistic stratospheric influence on the CO_2 mixing ratio. Also, models could fail to simulate the CO_2 seasonal cycle correctly because they have not been run over a sufficiently long time period to allow for stratosphere-troposphere exchange. We find that model runs at least 3 years are needed for surface CO_2 to be transported to the stratosphere and back to the troposphere at middle and high latitudes. *In situ* observations of CO_2 mixing ratios in the upper troposphere and satellite measurements of the column averaged CO_2 mixing ratios with accuracies around 1.0 ppmv or better would place strong constraints on the transport processes, especially the troposphere-stratosphere exchange. The validation of the model transport would increase our confidence in deducing CO_2 sources and sinks on the global scale using 3-D models [*Eluszkiewicz et al.*, 2000; *Baker et al.*, 2006].

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