Global-scale transport of carbon dioxide in the troposphere

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[1] An atmospheric transport model was used to examine the roles of variously scaled atmospheric transport processes (Lagrangian mean motions, large-scale eddies, and parameterized vertical diffusion and convective transport) in the spatiotemporal distributions of tropospheric carbon dioxide (CO2). The mean and eddy transports were analyzed using the mass-weighted isentropic zonal mean. We found several differences in the dominant transport processes for tropospheric CO2 distributions between the extratropics of both hemispheres and the tropics. (1) In the northern extratropics in boreal autumn to spring, CO2 emitted by anthropogenic and biospheric sources is uplifted and dispersed through quasi-isentropic eddy mixing associated with baroclinic waves and accumulates in the extratropical low-isentropic troposphere (in “cold pocket” below 300K). (2) High-CO2 air is transported from the northern extratropics into the tropics through low-level mean meridional flow. It is uplifted together with CO2 emitted by tropical vegetation through deep convection and diabatic eddies in the tropics during boreal winter to spring. (3) During summer at the northern midlatitudes, the low mixing ratio of CO2 produced by biospheric uptake is uplifted into the upper troposphere by convection and is strongly isolated from the lower latitudes. (4) The CO2 emitted in the Northern Hemisphere and the tropics is transported into the Southern Hemisphere via the tropical upper troposphere due to eddy mixing during boreal winter to spring and mean divergent flow of Hadley circulation during boreal summer. (5) In the Southern Hemisphere, an upward gradient of CO2 forms by upper tropospheric southward advection during boreal spring-autumn.


1. Introduction

[2] Observational studies have investigated long-term temporal and spatial variation in atmospheric CO2 near the Earth’s surface, with a main focus on the carbon cycle [e.g., Conway et al., 1994; Francey et al., 1995; Keeling et al., 1994]. Aircraft measurements and balloon observations have shown distinct variation in the upper air CO2 [e.g., Nakazawa et al., 1991; Hoor et al., 2004; Engel et al., 2006]. The tropospheric CO2 concentration exhibits large seasonal variation in the Northern Hemisphere (NH) extratropics, mainly because of net carbon release in winter and strong uptake in summer by land vegetation. This seasonal variation weakens toward the tropics and is largely absent in the Southern Hemisphere (SH). The amplitude of the seasonal variation in CO2 concentration generally decreases with height throughout the troposphere, with a phase lag in the upper troposphere compared to the lower troposphere; in the NH extratropics, this phase lag is about 1–2 months [Nakazawa et al., 1991]. Because CO2 is chemically inert in the troposphere, an understanding of variation in upper air CO2 can help in understanding not only the carbon cycle, but also atmospheric transport processes.

[3] Atmospheric transport processes at various scales influence the variation in chemical tracer distributions in the troposphere. Global-scale tropospheric transport processes have been investigated using Lagrangian analyses [Kida, 1983; Stone et al., 1999; Bratseth, 2003], which have suggested that the global troposphere can be divided into three parts: the tropics, NH extratropics, and SH extratropics [Plumb and Mahlman, 1987; Eruchimova and Bowman, 2006]. Transport within each region is rapid, whereas transport across the boundary around the subtropics is relatively slow. Plumb and Mahlman [1987] described the following routes for transport from the midlatitudes of one hemisphere to the other: lower tropospheric advection into the intertropical convergence zone, upward advection and diffusion in the tropics, and poleward advection into the upper troposphere of both hemispheres. They also indicated that the timescale for exchange between the high latitudes of the two hemispheres is limited by the timescale of the...
transport into the tropics across the subtropics, rather than by the transport across the tropics.

[4] The stirring mechanisms differ in the tropics and extratropics. In the extratropical troposphere of both hemispheres, air parcels disperse relatively rapidly because of large-scale eddy motions that force the vertical and horizontal extents of the tracer motion [Stone et al., 1999; Wang and Shallicross, 2000; Bowman and Erukhimova, 2004]. In particular, during a baroclinic-wave life cycle, tracers released in the boundary layer are transported into the free troposphere, upward and poleward along the warm conveyor belt (a rising stream of wet and warm air located in warm side of the cold front). Within the tropics, the overturning Hadley circulation disperses air. Even though the Hadley circulation varies considerably with the seasonal cycle, the transport barriers in the subtropics persist during all seasons [Bowman and Erukhimova, 2004].

[5] Convection also plays an important role in transporting atmospheric compositions in the troposphere. Moist convection moves polluted air from near the surface to the upper troposphere. Hess [2005] reported that lower tropospheric tracers are lofted to the middle and upper troposphere by convective and nonconvective processes. The interplay between moist convective and nonconvective transport (by large-scale circulation) explains the general aspects of the global tropospheric distribution of trace species. There is a barrier to the mixing of convectively processed air between the tropics and extratropics, and there is fairly rapid dispersion of air within the tropics. The mixing barrier between the tropics and extratropics extends approximately parallel to the 310 K isentropic surface [Bowman and Carrie, 2002]. In addition, small-scale transport via vertical diffusion acts to propagate variation in CO2 to the upper levels within the planetary boundary layer (PBL).

[6] Atmospheric transport models have been used to investigate the relative importance of transport processes at various scales to the CO2 distribution, such as by Lagrangian mean motions, large-scale eddies, and small-scale transport processes via convection and vertical diffusion [e.g., Strahan et al., 1998; Kawa et al., 2004; Tiwari et al., 2006]. Small-scale (or subgrid-scale) processes are generally parameterized in global transport models because of the insufficient resolution of current models. Understanding the atmospheric transport processes will provide important information to improve transport models, and realistic model transport would allow for unbiased estimation of surface CO2 fluxes using inversion or assimilation techniques. However, in previous work, the relative contributions of various transport processes to the global tropospheric CO2 distribution have not been well quantified. Major problems that preclude better understanding include the complicated estimation of meridional transport via large-scale eddy motions. Most analyses have also had difficulty in expressing CO2 transport near the Earth’s surface because of incomplete representation of the lower boundary condition, which hinders accurate budget analysis of tropospheric CO2.

[7] Here, in order to help understanding of atmospheric CO2 variation, we quantitatively describe the global-scale transport processes of tropospheric CO2 based on detailed transport analysis in models. The transport analysis is based on the mass-weighted isentropic zonal means, allowing the accurate analysis of Lagrangian mean motions and large-scale eddies [Miyazaki and Iwasaki, 2005]. In section 2, we briefly describe the global transport model and compare some representative modeled and observed CO2 distributions. In section 3, we introduce the analysis method for global-scale CO2 transport processes and discuss the zonal mean CO2 distribution in different coordinates. In section 4, we show the seasonal variation in the tropospheric CO2 distribution and present the results of the transport analysis. We conclude by summarizing the implications of the results.

In a companion study (K. Miyazaki et al., Formation mechanisms of the latitudinal CO2 gradients in the upper troposphere over the subtropics and tropics, submitted to Journal of Geophysical Research, 2008), we examine the evolution mechanisms of the latitudinal CO2 gradient around the Equator and NH subtropics [Miyazaki et al., submitted manuscript, 2008].
The nudged AGCM allowed the reproduction of past meteorological fields and the performance of transport calculations realistically at short time steps. The time step for integrating the transport calculation was 10 min. The nudging relaxation time was optimized and set to 1 d for wind and 5 d for temperature to reproduce the mean-meridional circulation for the objective analysis with reduced temperature bias in the nudged GCM [Miyazaki et al., 2005].

2.3. Simulation Description

A 4-year simulation of 2000–2003 was performed for two sets of surface fluxes to confirm the dependence of surface flux data on the simulated atmospheric CO2 variation. The control (CTL) simulation used surface fluxes due to anthropogenic emissions (annual mean [Brenket, 1998]) and seasonally varying atmosphere-biosphere fluxes calculated using an ecosystem model based on the Carnegie-Ames-Stanford approach (CASA [Randerson et al., 1997]) and atmosphere-ocean fluxes obtained from Takahashi et al. [2002]. The second set of surface fluxes (CYC simulation) was obtained from an atmospheric inversion model setup [Gurney et al., 2004] that uses a network of 87 stations [Patra et al., 2005]. Figure 1 shows the latitudinal distribution of total surface CO2 fluxes (zonal means). The northern middle and high latitudes, approximately 45°–70°N, exhibit large seasonal variation in surface CO2 fluxes, with a strong source during autumn to spring and a strong sink during summer. Distinct seasonal variation in surface CO2 fluxes is also present at low latitudes, roughly 25°S–25°N, with net emission during winter to spring and net uptake during summer to autumn. Biospheric components dominate the seasonal variation in CO2 flux at low latitudes. At southern midlatitudes, surface fluxes are negative throughout the year because of uptake by the Southern Ocean. The surface fluxes in the CTL and CYC simulations are generally in good agreement, except for stronger sinks at the midlatitudes of the NH (SH) in July (January) in the CYC simulation.

The initial distribution of the CO2 mixing ratio was set to be globally constant and a 5-year spin-up run was performed prior to the simulation of the 2000–2003 analysis period. A 5-year spin-up period was thought to be sufficient to simulate the CO2 distribution realistically in the troposphere and lower stratosphere, considering that the age of air in the lower stratosphere (about 4–5 years [e.g., Bischof et al., 1985; Boering et al., 1996]) is less than the spin-up period (the age of air is defined as the transport time of lower tropospheric air to the upper layers). Our transport analysis used 3-hourly data to include the influence of short-term atmospheric transient disturbances.

2.4. Validation

Simulations of CO2 using the CCSR/NIES/FRCGC transport model have been analyzed under a multimodel intercomparison program called the TransCom continuous experiment. As described by (P. K. Patra et al., TransCom model simulations of hourly atmospheric CO2: Analysis of synoptic scale variations for the period 2002–2003, submitted to Global Biogeochemical Cycles, 2007), the CCSR/NIES/FRCGC transport model is capable of representing synoptic-scale variation in the surface CO2 mixing ratio fairly realistically. The comparison here was to validate seasonal and latitudinal variation in simulated CO2 near the Earth’s surface and in the upper troposphere. The CO2 distributions obtained by the CTL and CYC simulations were compared with aircraft observations over the western Pacific in the upper troposphere (Figures 2c, 2e, and 2g). Observational data were obtained from flask samples taken during Japan Airlines (JAL) commercial flights between Narita, Japan (35.46°N, 140.23°E), and Sydney, Australia (33.58°S, 151.11°E), with an interval of 2 weeks [Matsueda et al., 2002]. Air samples were collected at cruising altitudes of 8–13 km above the ground surface. Modeled and observed surface CO2 mixing ratios were also compared at a Pacific site (Figures 2b, 2d, and 2f). The surface observational data were obtained from ship- and ground-based measurements [GLOBALVIEW-CO2, 2007]. Figure 2a shows the observational site locations. The mid-Pacific Ocean is located far from strong source and sink areas of the continental regions and is considered to be favorable for
validating the model results, with a focus on atmospheric transport processes.

[13] Figure 2 shows modeled and observed time-latitude cross-sections of the CO2 mixing ratio for comparison. Both the CTL and CYC simulations using the transport model capture fairly well the salient features of surface and upper tropospheric observations. Fossil fuel emissions, mainly located in the NH midlatitudes, are observed as elevated atmospheric CO2 concentrations overlaid on the CO2 flux signal of the terrestrial biosphere that dominates...

Figure 2. Time-latitude cross-section of the CO2 volume mixing ratio (shaded colors, in ppm) in the Western Pacific. The left panels show the CO2 concentration near the surface obtained from (b) ship- and ground-based measurements, and the (d) CTL and (f) CYC simulations averaged between 170°E and 120°W. The right panels show the CO2 in the upper troposphere obtained from (c) JAL flight observations at 10500 m, and the (e) CTL and (g) CYC simulations averaged from 300 to 260 hPa at 146°W. The data collected by JAL aircraft have been described by Matsueda et al. [2002]. The observational sites used are plotted in Figure 2a in which blue represents surface observations and red represents aircraft observational points.
the seasonal variation. In the NH, the CO2 mixing ratio in the upper troposphere increases from November to March and decreases from May to September, with a maximum in May and a minimum in September. The CO2 mixing ratio in the upper troposphere has a smaller seasonal-cycle amplitude with a phase delay compared to that near the Earth’s surface. The variation in CO2 in the NH extratropics propagates toward the tropics with decaying amplitude. Both the model and observation results show large latitudinal CO2 gradients around the Equator and NH subtropics in the upper troposphere, indicating an existence of a barrier to meridional transport. The amplitude of variation in CO2 is much lower in the SH than in the NH, especially at the surface.

[14] Compared to the CTL simulation, the CYC simulation showed better agreement with the observations, particularly in the SH. The CTL model simulation slightly underestimated the CO2 mixing ratio both at the surface and in the upper troposphere in the SH. This underestimation arises from stronger carbon uptake by the SH than the NH (Figure 1). We can thus use the results of the CYC simulation for the transport analysis. However, the two simulations showed very similar transport characteristics because of their common circulation field and overall agreement in surface flux distribution.

3. Methodology for the Transport Analysis

[15] Zonal mean transport analysis in a meridional plane provides a comprehensive survey of global-scale and long-term variation in CO2. In the middle to upper troposphere, the CO2 distribution is almost zonally symmetric and is substantially controlled by meridional and vertical motion (see section 5.1). Three-dimensional analysis for a specific event (at a specific time and place) can provide a simple explanation of the atmospheric transport process; however, transient (local) atmospheric flows strongly limit the generality of the understanding. Atmospheric chemical constituents are transported globally by mean-meridional circulation and large-scale eddies with scales greater than several thousand km, both of which are associated with atmospheric wave motions. The classic approach to studying these transport processes is the use of the Eulerian mean state in which the zonal average is taken at constant pressure surface; however, the Eulerian mean provides controversial insight into atmospheric transport problems. The Eulerian mean flow gives indirect circulation related to Stokes drift [Matsuno, 1980] (Figure 3a), which is inconvenient for understanding the meridional transport. In transformed Eulerian mean (TEM) analysis [Andrews and McIntyre, 1976], the tropical circulation is similar to the conventional Eulerian mean, whereas the extratropical indirect circulation of the Eulerian mean description (Ferrel circulation) is replaced by poleward direct circulation. Thus the tropospheric mean-meridional transport of the TEM analysis is caused by thermally induced Hadley circulation at low latitudes and by direct circulation due to baroclinic waves (e.g., synoptic-scale weather disturbance) in the extratropics (Figure 3b). Even though TEM analysis provides a sophisticated understanding of Lagrangian transport characteristics, it cannot discern the transport by eddy motion; in addition, the mass-stream function of the Lagrangian mean circulation intersects the lower boundary because of the incomplete representation of lower boundary conditions.

3.1. Description of Mass-Weighted Isentropic Zonal Means Analysis

[16] Most of the disadvantages in the conventional Eulerian and TEM analyses can be avoided by using mass-weighted isentropic zonal means (MIM) analysis [Tung, 1982; Iwasaki, 1989; Miyazaki and Iwasaki, 2005, 2008; Miyazaki et al., submitted manuscript, 2008], which accurately separates the meridional tracer transport into mean transport by Lagrangian-mean circulation and eddy transport by diffusive motions. In the MIM analysis, zonal average is taken on constant isentropic surface with mass weighting for all conservation parameters to express conservative nature of air mass and constituents. We use the isentropic zonal mean pressure for the vertical coordinate. MIM analysis is also capable of expressing lower-boundary and nongeostrophic effects on the mean meridional transports. The lower boundary effect facilitates inclusion of strong mean equatorward flow near the Earth’s surface (Figure 3c). The consideration of mass weighting for the zonal mean mixing ratio of constituents also ensures atmospheric tracer conservation. These criteria are important for accurate budget analysis of tropospheric CO2 in connection to different transport components.

[17] We analyzed global CO2 variation using a tracer continuity equation based on the MIM analysis, whereby the time derivative (tendency) of CO2 mass-mixing ratios contributed by mean transport, eddy transport, PBL mixing, and convective transport can be represented as

\[
\frac{\partial \text{CO}_2}{\partial t} = \frac{\nu}{a} \frac{\partial \text{CO}_2}{\partial \phi} - \frac{w_i}{\partial z_1} \frac{\partial \text{CO}_2}{\partial z_1} - \frac{1}{a \cos \phi} \frac{\partial (\text{CO}_2 \nu') \cos \phi}{\partial \phi} \\
- \frac{1}{\rho_0} \frac{\partial (\text{CO}_2 w_i')}{\partial z_1} + \frac{\partial \text{CO}_2}{\partial t} (PBL) \\
+ \frac{\partial \text{CO}_2}{\partial t} (\text{convection}),
\]

where \( \text{CO}_2 \) is the CO2 mixing ratio, \( z_1 \) is the log pressure coordinate, \( t \) is time, \( a \) is Earth’s radius, \( \nu \) is meridional wind, \( w_i \) is vertical wind, \( \phi \) is the latitude, and \( \rho_0 \) is the reference density. The overbars and asterisks denote the isentropic zonal mean and normalized mass weighting, respectively. Eddies are defined as departures from the mass-weighted zonal means. Eddies expressed as the primes are defined as departures from the mass-weighted zonal means. First and second (or third and forth) terms on the right-hand side of equation (1) represent the CO2 tendency by mean (or eddy) flux convergences.

[18] Air moving equatorward behind a cold front tends to have potential temperatures lower than the zonal average [Bowman and Carrie, 2002], and much of the equatorward branch of the mean overturning circulation occurs below the mean potential temperature of the Earth’s surface [Held and Schneider, 1999]. To represent this situation in MIM analysis, the surface pressure on the isentropic surfaces was set to be the surface pressure when isentropic lines intersected the ground surface; the local mass weight becomes zero at
these longitudes. This allowed us to represent accurately CO2 transport via lower tropospheric flow. Similarly, eddy transport flux becomes zero at the lower boundary because the eddy correlation vanishes there [Tanaka et al., 2004]. Further details on the MIM transport analysis have been reported by Miyazaki and Iwasaki [2005].

3.2. CO2 Profiles in Different Coordinate Systems: The Vertical Coordinate

Comparison of the zonal mean CO2 mixing ratio between different coordinate systems provides insight into atmospheric motion related to diabatic and diffusive processes. The CO2 distributions differ considerably if
expressed by the pressure coordinate, in which the zonal average is taken for the constant pressure surface, or by the isentropic coordinate, in which the zonal average is taken for the constant potential temperature surface (Figure 4). In the NH during winter, the CO2 mixing ratio generally increases with latitude in the pressure coordinate, but decreases with latitude in the isentropic coordinate. Compared to the pressure coordinate, the isentropic coordinate exhibits a smaller latitudinal CO2 gradient throughout the NH extratropics, suggesting the occurrence of adiabatic mixing that diffuses CO2-rich air from the middle to high latitudes along isentropic surfaces. Around the NH sub-tropics (25°N–30°N) during winter (in January), both the pressure and isentropic coordinates show large latitudinal gradients in the CO2 mixing ratio, but with opposite direction. The large latitudinal CO2 gradient in the isentropic coordinate implies that diabatic heating or cooling processes create a cross-isentropic CO2 slope at low latitudes. The diabatic transport process also causes upward CO2 slope in the NH midlatitudes during summer. In the SH during winter, the latitudinal CO2 gradient reaches its minimum around the 300 K isentropic surface. The location of the minimum gradient approximately corresponds to the lower boundary of the “middle world” (i.e., where isentropic surfaces do not intersect the ground surface [Hoskins, 1991]). This shows that isentropic mixing (mixing along local isentropic surfaces) is activated between the lower troposphere at low latitudes and the upper troposphere at high latitudes along the boundary between the middle world and lower world (i.e., where isentropes intersect the ground surface).

[20] The MIM analysis uses the mass-weighted isentropic zonal mean for the mixing ratio of CO2. Using the isentropic zonal mean, the CO2 concentration is evaluated at lower (higher) altitude in cyclonic low-pressure (anti-cyclonic high-pressure) systems than in a simple Eulerian mean. For example, in the lower troposphere of the SH midlatitudes during summer, the zonal mean CO2 mixing ratio is lower using the MIM analysis than using the Eulerian mean because the potential temperature is lower than the Eulerian zonal average in anticyclonic systems over the Southern Ocean (a strong sink area for atmospheric CO2).

[21] The time average for the surface sigma level (i.e., an average taken for the surface at a constant distance from ground level) allows for direct comparison of the surface CO2 concentration between the modeled and ground-based measurements. The difference in the zonal mean CO2 mixing ratio between the sigma-coordinate and pressure-coordinate systems can be negligible. An exception is the NH midlatitudes, where some differences arise from significant temporal and longitudinal variation in pressure and surface CO2 fluxes.

3.3. CO2 Profiles in Different Coordinates: The Latitudinal Coordinate

[22] Changing the latitudinal coordinate from the ordinary geographical latitude to the equivalent latitude also deforms the CO2 distribution. In the ordinary geographical coordinate system, both dissipative wave motions and apparent mixing due to conservative wave motions reduce the zonal-mean meridional CO2 gradient; the latter does not cause actual mixing. The use of the equivalent latitude coordinate allows the separation of the influences of conservative waves from those of dissipative waves on the zonal mean tracer field [Butchart and Remsberg, 1986; Nash et al., 1996]. Thus tracer distributions in the equivalent latitude coordinate system represent the tracer edge associated with dissipative wave motions and the mixing barrier. The effect of changing the latitudinal coordinate is generally small for the tropospheric CO2 distribution (Figures 4g and 4h), unlike for stratospheric tracers, suggesting that the latitudinal CO2 gradient in the ordinary geographical coordinate is not strongly affected by conservative wave motion and that tropospheric eddy CO2 transport mostly reflects actual mixing by dispersive motion. However, the CO2 distribution around the NH sub-tropics shows some disagreement between the two latitudinal coordinate systems during winter. The difference implies the occurrence of conservative Rossby wave motion around the subtropical westerly jets, as indicated by Scott and Cammas [2002]. For Lagrangian transport analysis, the use of an equivalent latitude-potential temperature coordinate is preferable [Hoskins, 1991]; however, this type of coordinate is not useful for showing the lower tropospheric CO2 distribution because of rapid changes in potential vorticity (PV) and in the CO2 mixing ratio along the PV contour, as well as because of active diabatic processes.

4. Meridional CO2 Distribution

4.1. Seasonal Variation

[23] Figure 5 shows meridional distributions of the zonal mean CO2 mixing ratio for 4 months. The largest variation in the zonal mean CO2 mixing ratio is found at 50°–60°N at the surface, with the maximum concentration in spring and the minimum concentration in summer and the NH extratropical troposphere charged with CO2-rich air during late winter and spring. In winter and spring, isolines of the CO2 mixing ratio are almost along the isentropes, and the CO2 concentration increases with latitude on the constant pressure surface in the middle and upper troposphere. The clear correspondence between the CO2 slope and isentropes (both slope poleward and upward) indicates rapid mixing along isentropic surfaces in the NH extratropics. By contrast, air masses with high CO2 in the extratropical lower troposphere are slowly transported to low latitudes.

[24] Both the CO2 slope and isentropes are steeper in the sub-tropics than in the extratropics in the NH during winter and spring. Large latitudinal CO2 gradients are located around 30°N in the lower troposphere and 40°N in the upper troposphere. The large CO2 gradients are roughly collocated with the transport barrier around the subtropical troposphere that extends from near 15°N at 900 hPa to 40°N at 400 hPa, approximately parallel to the 310 K isentropic surface [Bowman and Carrie, 2002]. The CO2 mixing ratio tends to largely decrease at the midlatitude NH during summer because of carbon uptake by land vegetation. This whole tropospheric summertime rapid decrease has been confirmed by aircraft observations over Siberia [Nakazawa et al., 1997]. The reduction in the CO2 mixing ratio at the NH midlatitudes during summer forms large latitudinal CO2 gradients around 40°N and 70°N. Cross-isentropic CO2 isolines at the NH midlatitudes imply that isentropic mixing of CO2 is significantly absent during summer.
Figure 4. Zonal mean CO$_2$ mixing ratio in the geographical latitude-pressure coordinate (upper panels), geographical latitude-mass-weighted isentropic zonal mean pressure coordinate (upper middle panels), geographical latitude-potential temperature coordinate (lower middle panels), and equivalent latitude-potential temperature coordinate (lower panels), averaged in January (left panels) and July (right panels) for 2000–2003. The contour intervals are 1 ppm.
autumn, the CO2 mixing ratio becomes almost constant in the NH extratropics throughout the troposphere.

[25] Seasonal variation in the CO2 mixing ratio in the NH extratropics weakens toward the tropics in the troposphere (Figure 5). The CO2 mixing ratio shows a large latitudinal gradient in the tropical troposphere during NH winter and spring. In contrast to the NH, the CO2 mixing ratio generally increases with height in the SH troposphere. These variations have been reported often by aircraft observations over the region of Australia and New Zealand [Pearman and Beardsmore, 1984]. Another distinct feature of the CO2 distribution in the SH is locally high concentrations at low latitudes (around 30°S to the Equator) during SH spring.

[26] Relatively weak vertical transport and mixing in the stratosphere compared to the troposphere cause sharp changes in vertical CO2 gradients near the tropopause. Aircraft observations have commonly exhibited the rapid changes in CO2 concentration near the extratropical tropopause [Hoor et al., 2004]. The strong isolation of the stratospheric and the tropospheric air controls the strength of the vertical CO2 gradient around the tropopause, particularly as associated with the tropopause inversion layer [e.g., Birner, 2006]. CO2 phase propagation related to stratosphere-troposphere exchange [e.g., Holton et al., 1995; Boering et al., 1996], and rapid poleward transport in the lowermost stratosphere [e.g., Hitchman et al., 1994].

4.2. Longitudinal Variation

[27] Eddy CO2 fields calculated from mean deviation from the zonal mean \( \left| \left[ CO_2 \right] - \left[ CO_2 \right]_{zonal} \right| \) can be seen as a measure of longitudinal variation in CO2 concentration. Figure 6 shows large-eddy CO2 fields in the lower troposphere, particularly in the NH extratropics. Variation in surface fluxes, PBL mixing, and near-surface local circulation is responsible for the lower tropospheric eddy fields. The eddy CO2 fields in the middle and upper troposphere are much smaller than those in the lower troposphere. This confirms that meridional and vertical motion together with surface flux variation substantially control the global CO2 distribution in the free troposphere. However, even in the free troposphere, the eddy CO2 field is large at low latitudes in the summer hemisphere and in the NH midlatitudes during summer compared to other latitudes (>0.4 ppm). These free tropospheric eddies are mainly caused by con-

Figure 5. Latitude-pressure distribution of the zonal mean CO2 mixing ratio (black contour lines with shaded color) and selected potential temperature (black lines) are shown for (a) January, (b) April, (c) July, and (d) October, averaged over 2000–2003. The contour interval is 1 ppm for the CO2 mixing ratio and 20 K for the potential temperature. The tropopause, defined by the temperature lapse rate, is indicated by a red line.
vective transport over the Inter-Tropical Convergence Zone (ITCZ) and NH midlatitude continental regions (Eurasian continent and North America). The eddy CO2 field is also large around the NH subtropics during NH summer. This distribution may result from Rossby wave motion and deep Asian monsoon circulation that effectively carry low-level air into the upper troposphere and lower stratosphere [e.g., Park et al., 2007]. The longitudinal CO2 variation, particularly associated with monsoon circulations, will be further discussed by Miyazaki et al. (submitted manuscript, 2008).

5. CO2 Transport Analysis

5.1. Mean Transport

The MIM analysis shows that the mean-meridional transport in the troposphere is caused by thermally induced Hadley circulation at low latitudes and by direct circulation due to baroclinic waves (e.g., synoptic-scale weather disturbance) in the extratropics (see, Figure 3c). The cross-isentropic Lagrangian-mean upward mass flux in the low latitudes corresponds to diabatic heating, whereas downward flux corresponds to diabatic cooling in the middle and high latitudes. The mean meridional transport fluxes of CO2 are directed poleward in the middle and upper troposphere and equatorward in the lower troposphere (Figure 7), causing an exchange of air between the tropics and extratropics. The mean-meridional circulation calculated from the MIM analysis captures a strong equatorward mean flow near the surface that effectively propagates variation in CO2 from the extratropics to the tropics. Mean poleward CO2 fluxes in the upper troposphere are much larger in the winter hemisphere than in the summer hemisphere. The poleward fluxes reach about 1300 ppm ms\(^{-1}\) at low latitudes of the NH and about 1400 ppm ms\(^{-1}\) at low latitudes of the SH in the upper troposphere during winter. Such fluxes quickly transport air from the tropics to the subtropics in the upper troposphere between approximately 300 and 100 hPa and from the subtropics to the extratropics in the middle and

![Figure 6. Latitude-pressure distribution of the mean deviation of the CO2 mixing ratio from the zonal mean (\(\left|\text{CO}_2\right| = \frac{\Delta_{\text{zonal}} \cdot \text{CO}_2}{2\pi} \right|\) in (a) January and (b) July for 2000–2003, with a contour interval of 0.2 ppm.](image)

![Figure 7. Latitude-pressure distribution of mean meridional CO2 transport fluxes in (a) January and (b) July for 2000–2003, with a contour interval of 150 ppm ms\(^{-1}\). Positive (or negative) values represent northward (southward) transport flux.](image)
5.1.1. NH Extratropics

5.1.2. Lower Troposphere

5.1.3. Tropics

Air in the extratropical lower troposphere (approximately below 2 km above the ground) is heated mainly by large-scale condensation and moves equatorward toward higher potential temperatures and is lofted into the upper troposphere in the tropics [e.g., Plumb and Mahlman, 1987; Bowman and Carrie, 2002]. The variation in low-level meridional flow follows the seasonal migration of the Hadley circulation in the tropics and of the wave-driven direct circulation in the extratropics. The low-level mean equatorward flow propagates variation in CO2 from the NH extratropics to the lower latitudes with a maximum tendency of about −8 ppm/month (Figure 8a). It generates a local maximum of the CO2 mixing ratio at NH low latitudes in the lower troposphere during NH winter and spring, together with CO2 emitted by land vegetation over central Africa, South America, and Southeast Asia and by the tropical eastern Pacific Ocean. Our analysis confirms that CO2 emitted in both the tropics (20°S–20°N) and the NH (20°–90°N) increases the CO2 concentration in the tropical lower troposphere, generating about 40% of the vertical CO2 gradient in the tropical middle troposphere during NH spring. In the SH, the CO2 tendency by lower tropospheric mean meridional transport is relatively small (less than 5 ppm/month) compared to that in the NH because of weaker surface fluxes.

5.1.3. Tropics

Air in the extratropical lower troposphere (approximately below 2 km above the ground) is heated mainly by large-scale condensation and moves equatorward toward higher potential temperatures and is lofted into the upper troposphere in the tropics [e.g., Plumb and Mahlman, 1987; Bowman and Carrie, 2002]. The variation in low-level meridional flow follows the seasonal migration of the Hadley circulation in the tropics and of the wave-driven direct circulation in the extratropics. The low-level mean equatorward flow propagates variation in CO2 from the NH extratropics to the lower latitudes with a maximum tendency of about −8 ppm/month (Figure 8a). It generates a local maximum of the CO2 mixing ratio at NH low latitudes in the lower troposphere during NH winter and spring, together with CO2 emitted by land vegetation over central Africa, South America, and Southeast Asia and by the tropical eastern Pacific Ocean. Our analysis confirms that CO2 emitted in both the tropics (20°S–20°N) and the NH (20°–90°N) increases the CO2 concentration in the tropical middle troposphere during NH spring. In the SH, the CO2 tendency by lower tropospheric mean meridional transport is relatively small (less than 5 ppm/month) compared to that in the NH because of weaker surface fluxes.

Figure 8. CO2 tendencies due to mean flux convergence \( \nabla \cdot \mathbf{v} = \frac{\partial \rho CO_2}{\partial z} + \frac{\partial \rho CO_2}{\partial y} \), in ppm month\(^{-1}\), shaded) averaged in (a) January, (b) April, (c) July, and (d) October for 2000–2003. The arrows indicate the mean CO2 transport fluxes.
transport via the Hadley circulation produces a negative CO2 tendency in the tropical upper troposphere, which persists through NH late autumn to early winter, with a maximum tendency of \(-3.6\) ppm/month in January. During these seasons, southward movement of the Hadley circulation induces upward transport of low-CO2 air and decreases the CO2 mixing ratio in the tropical upper troposphere. From NH early winter to summer, the mean transport tends to increase CO2 concentration in the tropical upper troposphere. The positive tendency results from the uplifting of high-CO2 air because of northward movement of the Hadley circulation. The mass stream function substantially intersects the CO2 slope around the Equator in equinox seasons (not shown), and mean transport produces a flattening effect on the CO2 slope in the tropical upper troposphere. The CO2 tendency by mean transport tends to be negative in the tropics during NH summer, reflecting the low-CO2 air transported mainly from the NH.

5.1.4. SH Extratropics

[32] Mean transport produces a positive CO2 tendency in the SH extratropics in contrast to the negative tendency in the NH extratropics. The mean southward flow propagates variation in CO2 from the tropics and increases the CO2 concentration in the SH extratropics. In particular, strong mean poleward flow causes rapid increases in the CO2 mixing ratio in the SH upper troposphere during SH summer and winter. The difference in the strength of the mean poleward flow between the upper and lower troposphere contributes to create an upward gradient of the CO2 mixing ratio in the SH extratropics.

[33] The transport model experiment indicates that surface fluxes in the NH largely contribute to the rapid increase in the CO2 mixing ratio in the SH upper troposphere, particularly during SH winter. The NH surface fluxes, from 20°N to 90°N, account for approximately 25% and 45% of the vertical CO2 gradient at 50°S at 400 hPa in April and July, respectively. Transport of CO2 from the NH surface into the SH upper troposphere occurs via the lower tropospheric mean meridional advection in the NH and tropical uplift. On this basis of aircraft observations of atmospheric CO (which has a strong source in the tropics) over the Southern Ocean, [Francey et al., 1999] similarly reported that the strong vertical gradient in CO at the NH midlatitudes during spring is induced by the middle tropospheric southward advection of air emitted by biomass burning in the tropical regions of southern Africa and South America. Upper tropospheric mean meridional flow is absent in summer and is responsible for the tendency for small summertime CO2 due to mean transport and the weak vertical CO2 gradient in the SH extratropics (Figure 8a).

[34] In contrast, in the SH lower troposphere, CO2 uptake by the Southern Ocean and land vegetation at low and midlatitudes of the SH (20°–50°S) largely decreases the CO2 concentration and increases the upward gradient of the CO2 mixing ratio, accounting for approximately 40% of the vertical CO2 gradient at 50°S at 700 hPa during SH autumn and spring. Around the SH subtropics, downward transport of high-CO2 air, excited by the downward branch of the Hadley circulation, increases the CO2 mixing ratio during SH autumn and winter. The high-CO2 air originates from the NH low latitudes through cross-equatorial mean meridional flow of the Hadley circulation.

5.1.5. Stratosphere

[35] The stratospheric mean tracer transport is driven by the Brewer-Dobson circulation. Its downward motion substantially deforms the stratospheric CO2 distribution, generally increasing the CO2 mixing ratio in the tropics and decreasing it in the extratropics in the lower troposphere. Downward transport fluxes are particularly strong in the winter hemisphere and intersect the extratropical tropopause. This largely decreases the CO2 mixing ratio at NH high latitudes in the upper troposphere and lower stratosphere during winter and spring (Figures 8a and 8b). The circulation makes a smaller contribution to variation in CO2 in the SH extratropics than in the NH extratropics because of the weaker velocity and smaller vertical gradient of the CO2 mixing ratio around the tropopause in the SH extratropics.

5.2. Eddy Transport

[36] Eddy transport processes consist of mixing via atmospheric disturbances (e.g., wave breaking events) and local circulation (e.g., monsoon circulation). The separation of eddy transport terms into diabatic and adiabatic components can aid in the understanding of transport mechanisms. The eddy vertical flux can be decomposed into diabatic and adiabatic parts with the diabatic heating rate \(\hat{\theta}^d\),

\[
\langle \text{CO}_2^* \rangle = \langle \text{CO}_2^* \rangle^d + \langle \text{CO}_2^* \rangle^a
\]

[37] Here, the eddy flux is parallel to the local isentropic surface for adiabatic processes. In addition, the CO2 tendency by eddy meridional transport is also separated into isentropic and cross-isentropic components as follows:

\[
\frac{1}{\rho_a} \left( \frac{\partial \langle \text{CO}_2^* v^\prime \rangle}{\partial \phi} \right)_{pi} + \frac{1}{\rho_a} \left( \frac{\partial \langle \text{CO}_2^* v^\prime \rangle}{\partial \phi} \right)_{pi} + \frac{1}{\rho_a} \left( \frac{\partial \langle \text{CO}_2^* v^\prime \rangle}{\partial \phi} \right)_{pi}
\]

Figures 9 and 10 show the resulting eddy CO2 transport due to eddy transport and its adiabatic and diabatic components.

5.2.1. NH Extratropics

[38] The NH extratropics exhibit distinct variation in eddy transport. Eddy transport flux is almost parallel to the local isentropic surface in the NH extratropics from autumn to spring, where both the eddy flux and isentropes slope poleward and upward. The eddy transport increases the CO2 mixing ratio in the NH extratropics from autumn to spring, with a maximum tendency of 4.5 ppm/month at NH high latitudes in the middle troposphere. The uplifting and dispersion of CO2 emitted by anthropogenic and biospheric sources are associated with baroclinic wave motion that causes significant meridional and vertical transport of tracers in the extratropical troposphere [Stone et al., 1999;
Uplifting transport ahead of the cold front of the warm conveyor belt in the extratropical cyclones has an important effect on the exchange of air between the boundary layer and upper levels, including the stratosphere [Stohl et al., 2003].

The adiabatic component dominates the eddy transport from autumn to spring in the NH (Figure 10a). The adiabatic eddy transport fluxes diverge at low and high latitudes and strongly converge at the midlatitudes in the NH during summer; this creates a large negative CO2 tendency at high latitudes (∼−5.5 ppm/month) and a positive tendency at midlatitudes (∼6.2 ppm/month) and acts to reduce the latitudinal CO2 gradient within the NH extratropics in the middle and upper troposphere. The positive tendency produced by diabatic eddy transport partly compensates for the strong reduction in CO2 through convective transport at the NH midlatitudes (see section 5.3.2 for discussion of the CO2 tendency due to convective transport), and the vertical eddy mixing dominated by diabatic processes tends to flatten the vertical CO2 gradient.

At NH low latitudes, the CO2 flux convergence due to eddy meridional transport acts to compensate for the reduction in CO2 by convective transport over the ITCZ during NH summer.

5.2.2. Tropics

Eddy mixing plays important roles in transporting CO2 from the NH into the SH in the tropical middle and upper troposphere during NH winter and spring (Figures 9a and 9b). Eddy transport produces a negative CO2 tendency...
at NH low latitudes and a positive tendency at SH low latitudes during these seasons. The adiabatic component dominates the interhemispheric eddy transport in the upper troposphere (Figure 10a), whereas the diabatic component drives significant vertical dispersion in the tropical middle and lower troposphere (Figure 10b). Although the eddy diffusion coefficient is relatively small in the tropics compared to the extratropics, the large latitudinal gradient in the CO2 mixing ratio causes the interhemispheric eddy mixing through the flux-gradient relationship. Miyazaki et al. (submitted manuscript, 2008) provides further discussion of interhemispheric transport processes.

5.2.3. SH Extratropics

The CO2 tendency due to eddy transport is much smaller in the SH extratropics than in the NH extratropics. It is particularly small in the upper troposphere, although eddy transport somewhat changes the lower tropospheric CO2 distribution. The CO2 emitted by the terrestrial biosphere in the region from approximately 30°S to the Equator is transported upward and poleward through eddy motion. This process somewhat increases the CO2 mixing ratio in the middle and lower troposphere during winter (~1 ppm/month). In contrast, during summer, downward and equatorward eddy transport carries air masses with relatively high CO2 mixing ratios from the pole to the midlatitudes (Figure 9a), somewhat increasing the CO2 mixing ratio over the Southern Ocean (approximately 6 ppm/month).

5.2.4. Stratosphere

Eddy transport also affects the lower stratospheric CO2 distribution. Rossby wave breaking brings quasi-adiabatic mixing between the tropics and extratropics in the lower stratosphere when westerlies are present [e.g., Waugh, 1996]. Poleward eddy transport decreases the CO2 mixing ratio in the tropics and increases it in the extratropics in the lower stratosphere. Meanwhile, quick transport of tropospheric air into the extratropical lower stratosphere is caused by eddy mixing around the NH subtropical tropopause during NH summer. Mixing across the subtropical tropopause is usually stronger in summer than in other seasons [Scott et al., 2003]; this variation is consistent with the seasonal variation in Rossby wave breaking events around the subtropical tropopause [Postel and Hitchman, 1999]. Deep Asian monsoon circulation also drives the cross-tropopause transport during summer [e.g., Park et al., 2007]. The observed high CO2 abundance in the lowermost stratosphere at the NH midlatitudes during summer [Aoki et al., 2003] is possibly associated with the cross-tropopause eddy transport around the subtropics.

5.2.5. Two-Dimensional Model Experiment

A two-dimensional (2-D) model that neglects CO2 transport via eddy motion was used to demonstrate the
importance of eddy transport for the tropospheric CO2 distribution. The 2-D model was constructed using the zonal mean transport equation of equation (1) in which CO2 is transported by the mean-meridional circulation and subgrid processes of vertical diffusion and convective transport and the eddy transport terms are neglected (note that the 2-D model with all transport terms including eddy motions produced a similar CO2 distribution to the 3-D model). The CO2 tendencies due to convection and vertical diffusion were obtained from the global transport model (3-D model) output. The comparison of the zonal mean CO2 distributions from the 3-D and 2-D models shows a significant difference in the NH extratropics (Figure 11). The 2-D model without eddy transport indicates a lower CO2 mixing ratio in the NH extratropics in the middle and upper troposphere than does the 3-D model; the difference indicates that there is significant transport via eddy motion causing strong uplift and dispersion of lower tropospheric CO2.

Figure 11. Latitude-pressure distribution of the zonal mean CO2 mixing ratio (black contour lines with shaded colors) obtained from the (a) 3-D transport model and (b) 2-D transport model, averaged during April and May for 2000–2003. In the 2-D model, the initial CO2 distribution was obtained from the zonal mean CO2 distribution of the 3-D model simulation on 1 April. The contour interval is 1 ppm. The arrows indicate eddy transport fluxes.

The 2-D model results confirm the significance of eddy motion to the interhemispheric CO2 transport from the NH into the SH during NH spring. In the 2-D model, the tropical CO2 distribution follows the overturning Hadley circulation. The implications of the 2-D model results agree...
well with the analysis of eddy transport terms made from the 3-D model.

5.3. Convective Transport and Vertical Diffusion

5.3.1. PBL Mixing

A significant spatial dependence in the relative contribution of subgrid and large-scale transport processes on the lower tropospheric CO2 mixing ratio. Vertical diffusion propagates the surface CO2 variation into the PBL, which has a large effect on the lower tropospheric CO2 distribution over continental regions, particularly during the summer. The CO2 tendency due to PBL mixing reaches up to approximately 600 hPa at the NH midlatitudes during summer, with the maximum value reaching approximately −28 ppm/month at 850 hPa (Figure 13). The negative CO2 tendency produced by the PBL mixing reflects the low CO2 mixing ratio emitted by the terrestrial biosphere. During winter, the CO2 tendency is restricted to a lower level compared to the level in summer because of the lower PBL height. Carbon release and uptake in central Africa and northern South America produce large seasonal variation in the lower tropospheric CO2 mixing ratio through vertical diffusion from the tropics to the SH subtropics. In the SH extratropics, uptake by the Southern Ocean and land vegetation over South America decreases the CO2 mixing ratio in the lower troposphere during summer in association with the strong vertical diffusion in summertime.

5.3.2. Convective Transport

Convective transport contributes significantly to the tropospheric tracer distribution [e.g., Gray, 2003; Erukhimova and Bowman, 2006]. The comparison of outgoing longwave radiation produced by the model and from satellite measurements confirms that the model captures well the main features of the cloud distribution and occurrence of convective clouds (not shown). Convective updraft and downdraft motion enhances the vertical transport of CO2 and creates a strong connection between the lower and upper troposphere in the tropics. The convective transport is particularly obvious over the ITCZ in the rising branch of the Hadley circulation (Figure 12a). Upward transport by convection largely increases the CO2 mixing ratio from approximately 300 to 100 hPa in the tropics during NH winter, whereas it causes the uplift of low-CO2 air into the middle and upper troposphere during NH summer. Convective activity also has a great effect on the CO2 distribution in the NH midlatitudes during summer, with convective transport producing a large negative CO2 tendency in the middle and upper troposphere in the NH midlatitudes (−13 ppm/month) by lifting low-CO2 air emitted by land vegetation over Eurasia and the NH areas of the Americas. At the same time, convective...
downdraft motion produces a positive CO2 tendency (approximately 6 ppm/month) at the NH midlatitudes in the lower troposphere, compensating for the reduction due to vertical diffusion.

[48] The effect of convective transport on the tropospheric CO2 distribution was assessed using a transport model experiment that neglects CO2 transport by parameterized convection. Figure 14 shows the difference (convective transport enabled minus convective transport) in the zonal mean CO2 mixing ratio between the simulations with and without convective transport for January and July. By including the CO2 transport by convection, the CO2 concentration increases (or decreases) in the NH (or SH) low latitudes in the middle and upper troposphere, thus making the latitudinal CO2 gradient around the Equator steeper. A large anomaly extends from the tropics to the extratropics in the upper troposphere in the NH winter. The extension of the anomaly is obvious in the poleward advection in the upper troposphere. A notable difference also occurs in the NH midlatitudes during summer; the zonal mean difference between the two simulations is approximately 0.6 ppm in the tropical upper troposphere in NH winter. The extension of the anomaly is obvious in the poleward advection in the upper troposphere. A notable difference also occurs in the NH midlatitudes during summer; the zonal mean difference between the two simulations is approximately 0.6 ppm in the tropical upper troposphere in NH winter. The extension of the anomaly is obvious in the poleward advection in the upper troposphere. A notable difference also occurs in the NH midlatitudes during summer; the zonal mean difference between the two simulations is approximately 0.6 ppm in the tropical upper troposphere in NH winter.

5.4. Conceptual Picture of Global-Scale CO2 Transport

[49] Our analysis of different transport processes provides a comprehensive view of global-scale variation in the tropospheric CO2 distribution (Figure 15). The dominant transport mechanisms of the tropospheric CO2 distribution differ between the extratropics and the tropics, as well as among seasons. Previously, Bowman and Carrie [2002] have illustrated a schematic diagram of tropospheric tracer transport from a Lagrangian-based approach.

5.4.1. NH Troposphere: Autumn to Spring

[50] Vertical mixing propagates surface CO2 variation into the PBL. The PBL mixing reaches up to approximately 600 hPa during summer at NH midlatitudes, and the PBL height is lower during winter. The CO2 emitted by anthropogenic and biospheric sources is further uplifted from the boundary layer into the free troposphere in the NH extratropics during autumn to spring through synoptic-scale eddy motion associated with baroclinic waves. The extratropical eddy transport fluxes are almost parallel to isentropic surfaces; these fluxes slope poleward and upward from the lower troposphere at low latitudes to upper troposphere at high latitudes. As a result of the quasi-isentropic eddy motion, the NH extratropical troposphere is charged with CO2-rich air during winter to spring where the CO2 mixing ratio increases with latitude at a constant pressure surface (or at constant altitude). Extratropical air masses are rarely diffused into low latitudes and remain within the extratropics, particularly within the cold pocket below approximately the 300 K isentropic surface. This is because of rapid changes in the thickness of the air layer below about 300 K isentrope around the subtropics. In the middle and upper troposphere, there is also a clear correspondence between large latitudinal gradients in the CO2 mixing ratio and isentropes around the NH subtropics, implying the occurrence of an eddy mixing barrier. The mean poleward flow propagates low-CO2 air from the tropics to higher latitudes and decreases the CO2 mixing ratio in the NH extratropics in the middle and upper troposphere. This partly compensates for the increase in the CO2 mixing ratio caused by eddy dispersion.

5.4.2. NH Troposphere: Summer

[51] The atmospheric CO2 concentration decreases rapidly in the NH extratropics during summer because of strong biospheric carbon uptake. Moist convection causes significant uplift of low-CO2 air and largely decreases the CO2 concentration in the middle and upper troposphere at the NH midlatitudes. The low-CO2 region produced by convective transport is strongly isolated from the lower latitudes as a result of summertime weak air exchange across the NH subtropics in association with summertime slow mean-meridional circulation and inactive wave activ-
This builds a large latitudinal gradient in the CO2 mixing ratio around 40°N. Down-gradient mixing via eddy motion partly compensates for the reduction in CO2 produced by convective transport at the midlatitudes and smoothes the CO2 distribution within the NH extratropics. The eddy CO2 transport substantially reflects diabatic processes during summer.

5.4.3. Lower Troposphere

Low-level mean-meridional circulation is an important pathway for the propagation of variation in CO2 from the extratropics in the winter hemisphere to the tropics and into the summer hemisphere. The mean-meridional advection effectively carries high-CO2 air from the NH extratropics into the tropics in the lower troposphere during NH
winter and is primarily responsible for a rapid increase in the CO2 mixing ratio in the tropical lower troposphere during NH winter to spring, along with CO2 release by the tropical oceans and vegetation. Low-level meridional flow is substantially absent and produces the small CO2 tendency throughout the hemisphere during summer. In contrast, in the SH, low-level meridional flow does not largely change the CO2 distribution because of weak surface CO2 fluxes year-round.

5.4.4. Tropics

[55] Air at low levels ascends mainly by moist convection in the tropical troposphere. The convective ascending causes the uplift of CO2 emitted in the tropics and the NH and increases the CO2 concentration in the tropical middle and upper troposphere, especially at the NH low latitudes during NH winter and spring. Vertical eddy transport excited by active diabatic processes also brings significant uplift in the tropics. The uplift magnifies the interhemispheric difference in the CO2 mixing ratio in the tropical middle and upper troposphere. These vertical transports make less of a contribution to the tropical CO2 distribution during NH summer than during other seasons because of the small CO2 tendency in the tropical lower troposphere through low-level mean-meridional flow.

[54] Effective transport of CO2 emitted in the NH into the SH occurs in the tropical upper troposphere. Thus the CO2 emitted in the NH extratropics is transported into the SH via low-level equatorward advection and tropical uplift. These transport pathways have been commonly described by Plumb and Mahlman [1987]. Our analysis further confirms the dominant mechanisms for interhemispheric CO2 transport in the tropical upper troposphere by cross-equatorial eddy fluxes during NH winter to spring and by mean southward flow via tropical Hadley circulation during NH summer.

5.4.5. SH Troposphere

[55] Variation in tropospheric CO2 differs greatly between the NH and SH. The CO2 mixing ratio increases with altitude from autumn to spring in the SH. The CO2 released by NH sources substantially increases an upward gradient in the CO2 mixing ratio in the SH middle and upper troposphere, especially during NH winter and spring, through effective interhemispheric transport in the tropical upper troposphere. In addition, air masses with relatively low CO2 mixing ratios remain within the SH extratropical cold pocket in the lower world and are strongly isolated from air masses at lower latitudes; the boundary between the two air masses stays near the 300 K isentropic surface. This also contributes to the production of an upward gradient of the CO2 mixing ratio in the middle and upper troposphere. In contrast, in the lower troposphere, carbon uptake by the Southern Ocean and land vegetation increases the upward CO2 gradient during SH winter. Around the SH subtropics, mean descent in the Hadley circulation and ascent by adiabatic eddy motion increase the CO2 mixing ratio in the middle troposphere during SH winter.

5.4.6. Lower Stratosphere

[56] In the winter hemisphere, the CO2 mixing ratio in the extratropical upper troposphere and lower stratosphere decreases due to the mean downward transport by the Brewer-Dobson circulation. The CO2 tendency by mean downward transport is larger in the NH than in the SH due to stronger descent and the larger vertical CO2 gradient around the tropopause. In contrast, intertropical eddy mixing tends to flatten the CO2 slope almost along isentropic surfaces in the lower stratosphere.

[57] Distinct cross-tropopause transport of CO2 from the troposphere into the stratosphere occurs around the NH subtropics during summer. Poleward eddy transport brings tropospheric high-CO2 air into the lowermost stratosphere around 35°–45°N. Rossby wave breaking and Asian monsoon circulation are possibly responsible for the cross-tropopause eddy transport fluxes.

6. Conclusions and Discussion

[58] We investigated global-scale CO2 transport processes in the troposphere using a transport analysis that uses a meridional transport equation based on mass-weighted isentropic zonal means. The meteorological fields and CO2 distributions were obtained from on-line global transport model driven by a nudged GCM. The transport model considered both grid-scale and subgrid-scale CO2 transport processes; subgrid processes included parameterized convective transport and turbulent mixing. The simulated CO2 distribution was compared with aircraft- and ground-based measurements over the Pacific. The comparison shows that the transport model captures the observed latitude-time structure of the CO2 mixing ratio fairly well both at the surface and in the upper troposphere.

[59] We examined the global-scale CO2 transport processes from estimations of Lagrangian-mean motion and large-scale eddies and obtained the following insights into tropospheric CO2 variation. (1) Eddy mixing brings the uplift and dispersion of CO2 emitted by anthropogenic and biospheric sources in the NH extratropics associated with baroclinic wave motion during autumn to spring. The eddy transport fluxes slope poleward and upward almost along isentropic surfaces. High CO2 mixing ratios particularly accumulate within the extratropical “cold pocket” (i.e., below the approximately 300 K isentropic surface) during winter to spring. A 2-D model simulation that neglected eddy CO2 transport was used to confirm the importance of eddy motion to the extratropical CO2 distribution. Extratropical CO2 is marginally diffused into the lower latitudes because of rapid decreases in the low-level air layer below the 300 K isentrope around the subtropics. The CO2 concentration in the NH extratropics in the middle and upper troposphere is somewhat decreased by the poleward-downward mean transport. (2) CO2 emitted in the NH extratropics is partly transported to lower latitudes through low-level mean-meridional flow. The mean-meridional flow effectively propagates variation in CO2 from the NH extratropics into the tropics and produces a high-CO2 region in the tropical lower troposphere during NH winter and spring. CO2 emitted by sources in the NH and tropics is uplifted by tropical convection and diabatic eddy motion at low latitudes. (3) In contrast, during summer, biospheric uptake decreases the atmospheric CO2 concentration in the NH extratropics. Deep uplifting by convective processes produces a low-CO2 region throughout the troposphere of the NH midlatitudes. (4) Cross-equatorial eddy flux causes the efficient interhemispheric transport of CO2 from the NH into the SH during NH winter and spring, whereas mean
southward flow in the tropical Hadley circulation dominates the interhemispheric transport in NH summer. (5) In the SH, by moving air from the tropics to the extratropics in the upper troposphere, the vertical CO2 gradient becomes upward during autumn to spring. The CO2 released in the NH largely contributes to the development of the upward CO2 gradient in the upper troposphere during winter to spring.

[60] In recent years, comparisons of the inversion results (to estimate surface CO2 flux) between the different transport models have characterized the uncertainty of atmospheric transport models [Denning et al., 1999; Gurney et al., 2004]. The accurate analysis of surface sources and sinks requires improvements in transport models, along with the expansion of the observation network [e.g., Patra et al., 2006]. Detailed analyses of upper air variation in CO2 help to elucidate model accuracies from the process-oriented perspective. Our analysis should also be helpful in understanding the column CO2 variation that will be observed by satellites such as the Greenhouse Gasses Observing Satellite (GOSAT) and the Orbiting Carbon Observatory (OCO). The seasonal variation in column CO2 is generally delayed in phase with less variability compared to that at the surface as a result of the time delays associated with vertical mixing and the greater mass of CO2 in the column [e.g., Olsen and Randerson, 2004]. At the same time, these satellite data will allow the validation of the CO2 distribution obtained from the atmospheric transport model. Further analysis using both satellite and model data will be required to improve the understanding of the transport processes and the carbon cycle.

[61] Although our study has provided a comprehensive understanding of tropospheric variation in CO2, a question still remains regarding how the latitudinal gradients in CO2 mixing ratios develop around the subtropics and equator. The relative contributions of surface flux and atmospheric circulation to the seasonal evolution of the latitudinal CO2 gradient are not completely understood. Furthermore, the mechanisms of the interannual variation in atmospheric circulation and its affect on the tropospheric CO2 distribution remain unclear. Further detailed transport analyses are needed to survey the process that controls the latitudinal CO2 gradients and long-term variation in CO2.

[62] Furthermore, it is possible that the fraction of the tropospheric CO2 distribution due to transport by parameterized processes varies with the model resolution, cumulus convection scheme, and diffusion coefficient. Atmospheric tracer transport simulation requires the accurate representation of multiple aspects of convective variability [e.g., Lintner et al., 2004]. The model used here somewhat underestimates the deep convection over the maritime continent region and shows a broad convergence zone in the ITCZ (not shown). In addition, Gray [2003] and Rind et al. [2008] indicated that estimates of transport are highly sensitive to model resolution and to the artificial diffusion required for model stability. The uncertainty in CO2 transport simulations must be reduced to refine the outcomes.

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