Satellite remote sounding of mid-tropospheric CO_2

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[1] Human activity has increased the concentration of the earth's atmospheric carbon dioxide, which plays a direct role in contributing to global warming. Mid-tropospheric CO_2 retrieved by the Atmospheric Infrared Sounder shows a substantial spatiotemporal variability that is supported by *in situ* aircraft measurements. The distribution of middle tropospheric CO_2 is strongly influenced by surface sources and large-scale circulations such as the mid-latitude jet streams and by synoptic weather systems, most notably in the summer hemisphere. In addition, the effects of stratosphere-troposphere exchange are observed during a final stratospheric warming event. The results provide the means to understand the sources and sinks and the lifting of CO_2 from surface layers into the free troposphere and its subsequent transport around the globe. These processes are not adequately represented in three chemistry-transport models that have been used to study carbon budgets.
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1. Introduction

[2] Annual trends and seasonal changes in the CO_2 concentration have been monitored in situ since the mid-1950s [*Keeling et al.*, 1989]. The Atmospheric Infrared Sounder (AIRS) instrument, orbiting earth on NASA's Aqua satellite in a sun-synchronous near-polar orbit since 2002, affords us the capability to retrieve daily CO_2 concentrations for the first time globally, over land, ocean and polar regions, daytime and nighttime and in the presence of clouds, with an accuracy better than 2 parts per million by volume (ppmv) (i.e., <0.5%) and without relying on a priori or background information [*Chahine et al.*, 2005]. We retrieve the mid-tropospheric CO_2, employing a set of 15 μm spectral channels that have peak sensitivities to CO_2 around 450 hPa (see auxiliary material[1] Figure S1). The spatial resolution of our CO_2 retrieval is 90×90 km^2 in the nadir. In this paper we first assess the accuracy of the AIRS CO_2 retrievals and then compare them to the output of chemistry transport models. Finally, we relate the features in the AIRS CO_2 distribution to large-scale atmospheric circulation and major surface sources.

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2. Comparison With *in Situ* Measurements

[3] An earlier study [*Chahine et al.*, 2005] compared the monthly seasonal variations of AIRS retrievals to Matsueda airborne measurements [*Matsueda et al.*, 2002] (the data are available at ftp://gaw.kishou.go.jp/pub/data/current/co2/event/eom999900.mri.am.fl.co2.nl.ev.dat) for the period between September 2002 and March 2004 and showed an agreement of 0.43±1.20 ppmv. In addition, we have carried out further comparisons with available collocated *in situ* observations from aircraft. We compare AIRS CO_2 with *in situ* aircraft flask measurements obtained at cruising altitudes between 9.8 km and 11.6 km during commercial flights between Australia and Japan [*Matsueda et al.*, 2002] and confirm the accuracy is better than 2 ppmv (see Figure S2). We have also compared the spatiotemporal variations of AIRS CO_2 across North America to Intercontinental Chemical Transport Experiment – North America (INTEX-NA) measurements during July 2004 (H. D. Singh et al., INTEX-NA: Intercontinental Chemical Transport Experiment-North America, 2002, available at http://cloud1.arc.nasa.gov/docs/intex-na/white_paper.pdf; data from the 2004 campaign are available at http://www-air.larc.nasa.gov/cgi-bin/arcstat/) [*Choi et al.*, 2008]. Variations as large as 2-3 ppmv appear in AIRS CO_2 and are supported by INTEX-NA. Similar spatial variation of free tropospheric CO_2 over the continental United States were observed in June 2003 by the CO_2 Budget and Regional Airborne Study: North America 2003 (COBRA-NA, metadata file file.txt, available at http://www.fas.harvard.edu/~cobra/, 2003) aircraft field campaign [*National Oceanic and Atmospheric Administration*, 2006].

[4] Figure 1 compares AIRS CO_2 retrievals with measurements by the upward viewing Fourier Transform Infrared Spectrometer (FTIR) at Park Falls Wisconsin (45.93 N, 90.45 W) for the period between July 2004 and March 2006 [*Lin et al.*, 2006]. The details for converting the FTIR measurements to mixing ratios are given by *Washenfelder et al.* [2006]. The data presented in Figure 1 are monthly averages of the FTIR measurements and that of AIRS retrievals collocated within a radius of 250 km over a 19 month time span. AIRS measurements are sensitive to the mid- and upper-troposphere but not to the near-surface layer. The FTIR is sensitive to the entire atmospheric column. Both sets of measurements capture the seasonal variation of CO_2, and the FTIR measurements exhibit greater amplitude due to their sensitivity to the near-surface CO_2 where the impact of the seasonal summer uptake by vegetation is more pronounced (lower

[1]Auxiliary materials are available in the HTML. doi:10.1029/2008GL035022.

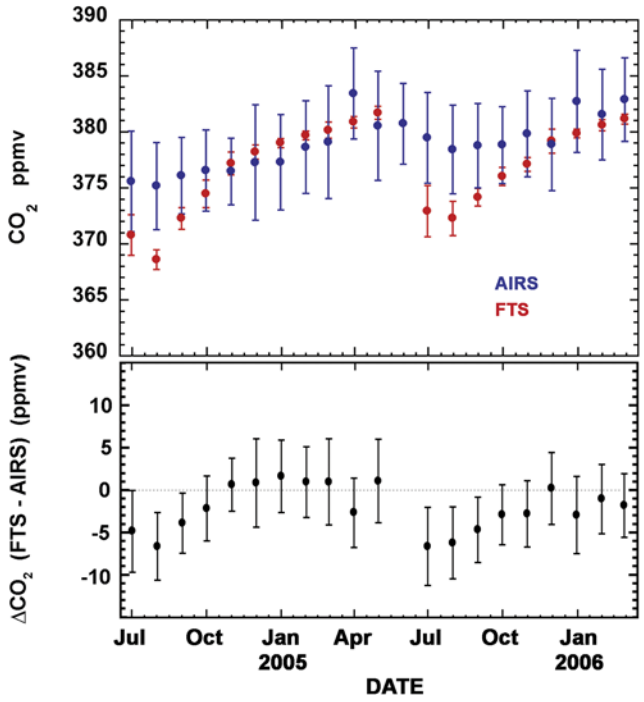

Figure 1. Seasonal variation of monthly average AIRS retrieved CO_2 within 250 km of Park Falls, Wisconsin compared to monthly average Park Falls Fourier Transform Spectrometer measured total column CO_2 and their differences.

panel). The AIRS CO_2 retrievals, which are obtained day and night and in the presence of clouds, are affected by tropospheric weather.

3. Comparison With Models

[5] In parallel with the observational evidence, we compare in Figure 2 the zonal averages of outputs of four Chemistry Transport Models (CTMs) (see auxiliary material) for April and July of 2003 and 2004 with the zonal averages of AIRS retrievals. The model profiles have been converted for comparison by applying the appropriate (tropical, mid-latitude or polar) weighting function of the AIRS CO_2 channels to calculate weighted averages and then averaging the results over all longitudes into 2 degree latitude bins. Significant differences between the AIRS observations and model results are apparent in all four panels.

[6] The spring-summer seasonal cycle of the northern hemisphere CO_2, i.e., the build up in the middle and high latitudes to a peak in April followed by a seasonal draw down in July by vegetation uptake, is readily apparent in the AIRS data. The 2003 cycle is more pronounced than that of 2004. Model CO_2 in the mid-latitudes of both hemispheres are systematically lower than AIRS retrievals by 3-5 ppmv in 2003. Similar deficits are apparent in the northern polar region in boreal spring of 2003, becoming smaller in boreal summer of 2003. The overall zonal variation of AIRS retrievals and model results is consistent in both hemispheres. However, the fine structure in the zonal variation of AIRS retrieved CO_2, usually appearing as enhancements in the extratropics, is not captured by any of the models. The origin of this fine structure is discussed later in this paper.

[7] We use data from 2003 in this study because significant work has been carried out covering this time period [*Chédin et al.*, 2003; *Crevoisier et al.*, 2004; *Engelen and McNally*, 2005; *Shia et al.*, 2006]. We now focus our analysis on the July 2003 global distribution of AIRS retrieved CO_2 and its comparison to CTMs. We have already seen in Figure 2 that AIRS retrieved CO_2 shows significant enhancements over the models in the mid latitudes of both hemispheres. To better understand their origin we turn our attention to the map of the retrieved CO_2 shown in the two panels of Figure 3. The top panel of Figure 3 shows AIRS retrieved CO_2 averaged over the month of July 2003 overlain by monthly averages of the National Center for Climate Prediction global reanalysis (NCEP2) 500 hPa geopotential height for reference. In general, the wind vectors run parallel to the geopotential height contours. It is readily apparent that the spatial variability of AIRS retrieved CO_2 is consistent with the large-scale circulation features in the pressure regime of peak AIRS sensitivity. The auxiliary material includes Animation 1[2], which shows the distribution and circulation of the mid-tropospheric CO_2 during July 2003.

[8] A striking feature in the top panel of Figure 3 is the greater than 3 ppmv spatial variability in the free troposphere. Global 3-D CTMs do not predict spatial variability of this magnitude. *Yang et al.* [2007] have also noted similar weak vertical mixing in current CTMs. The Goddard Earth Observing System (GEOS-Chem) simulations of CO_2 forced by sources and sinks shown in the bottom panel of Figure 3 display distributions that have much smaller spatial variability and lack the mid-latitude enhancements observed in the AIRS retrievals. Results from the Model for Ozone And Related chemical Tracers (MOZART-2) are similar to those from GEOS-Chem.

3.1. CO_2 Weather

[9] We note also that the AIRS retrieved mid-tropospheric CO_2 mixing ratios exhibit strong latitudinal and longitudinal gradients around 45 N, the location of the northern hemisphere (NH) mid-latitude jet stream. The distributions of the NCEP2 500 hPa geopotential heights shown in Figure 3 and the 500 hPa NCEP2 zonal winds suggest that the reduced concentrations of CO_2 north of the northern hemisphere mid-latitude jet stream are due to the combination of surface uptake by vegetation and vertical redistribution of air with low CO_2 concentrations from the stratosphere. The relatively high concentrations at 30 N to 40 N, south of the jet stream, correspond to the so-called NH mid-latitudes pollution belt [*Zhang et al.*, 2006]. In particular, the Southwest U.S. is affected by seasonal drought, which leads to vegetation stress and subsequently reduced photosynthesis. The unfavorable growing condition and emissions of CO_2 from fossil fuel combustion lead to enhancements of CO_2 across the Southwest U.S. Spatial gradients of 2–3 ppmv are apparent in the AIRS retrievals over the continental U.S.

[10] Over the western North Atlantic the higher CO_2 likely reflects northeastward transport of emissions from

[2] Animations are available in the HTML.

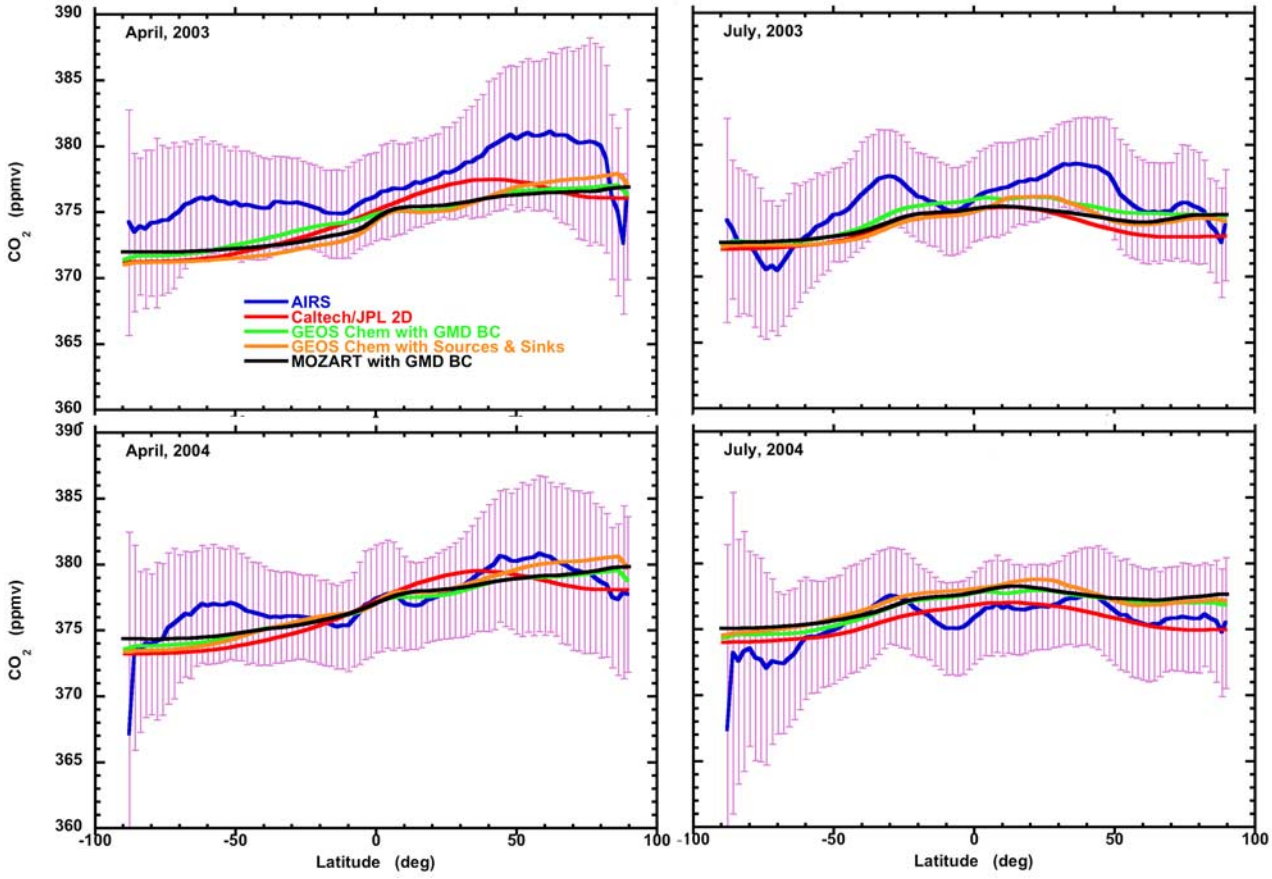

Figure 2. Zonal mean of AIRS retrieved CO_2 compared to models for April 2003, July 2003, April 2004 and July 2004. AIRS monthly average retrieved CO_2 (blue line) in 2 degree latitude bins averaged over all longitudes. The dashed vertical bars (purple) are the standard deviation of AIRS retrievals in each bin. Models, identified in legend of April 2003 graphic, are similarly averaged after convolving their CO_2 profiles with the appropriate (tropical, mid-latitude or polar) average weighting function of the AIRS CO_2 channels.

the southeast U.S. by warm conveyor belts, a dominant transport pathway for lifting pollution from surface into the middle and upper troposphere [*Stohl et al.*, 2002]. During the summer, the middle and upper troposphere over the Mediterranean is dubbed the 'global air pollution crossroads' with dominant influences from Asia and North America [*Lelieveld et al.*, 2002]. The higher CO_2 concentration over that region is a clear indication of long-range transport from North America to the west and from Asia to the south. *Li et al.* [2001] pointed out that transport of South Asia pollution via the tropical easterly jet leads to an ozone maximum in the middle troposphere over the Middle East. This long-range transport likely contributes to the high CO_2 concentrations over the Middle East and the Mediterranean. Continental outflow is also evident off the coast of East Asia.

3.2. Southern Hemispheric Circulation Patterns

[11] In the southern hemisphere (SH), the AIRS retrievals in Figure 3 exhibit a zone of relatively high CO_2 mixing ratios in the latitude band 30 S to 40 S. This zone of high CO_2 concentrations is within the SH subtropical storm tracks and the "convergence zone" cloud bands seen in these regions [*Hoskins and Hodges*, 2005]. This zone of high CO_2 likely indicates convective activity has lifted boundary layer air with higher concentrations of CO_2 into the free troposphere which becomes entrained in the SH midlatitude jet stream for subsequent rapid transport around the globe. The persistence of this zonal band of CO_2 is due to the zonal uniformity of the 500 hPa geopotential heights around the Southern oceans, which act as an atmospheric conveyor belt. In contrast to the NH, the strongest meridional temperature gradients in the midlatitudes are found in the SH summer season. Whereas the summer northern hemisphere (NH) storm-track activity is much weaker and shifts towards the pole, the SH activity is as strong in summer as in winter and shifts slightly towards the equator and with greater zonal symmetry [*Trenberth*, 1991]. The sources supplying this belt are discussed in the auxiliary material and shown in Animation 1.

[12] We have observed a region of decreased CO_2 concentration poleward of 60 S, whose origin we are still investigating.

3.3. Stratosphere-Troposphere Exchanges

[13] Stratosphere-troposphere exchange is expected to have a significant influence on upper tropospheric CO_2 at high latitudes [*Shia et al.*, 2006]. We should expect this

influence to be readily detectable in the AIRS CO_2 retrievals, due to their sensitivity to the middle and upper troposphere. We focus here on a stratospheric major final warming event over the northern polar region in April 2003. The World Meteorological Organization (WMO) criteria for stratospheric major warming are a reversal of the 10 hPa wind direction in the polar region and an increase of the stratospheric polar temperature. The top panel of Figure 4 shows the increase of AIRS retrieved 10 hPa temperature and reversal of NCEP zonal wind observed north of 60 N, which indicate that a major warming occurred on April 15, 2003.

[14] The polar vortex becomes greatly distorted following a major final warming and eventually breaks up. As a result, there is less downwelling of stratospheric air into the polar upper troposphere and more mixing with air from northern mid-latitudes [*Limpasuvan et al.*, 2004]. The transport of high concentration of CO_2 and low concentration of O_3 from the mid-latitudes to high-latitudes leads to an increase in concentrations of CO_2 and a decrease in the concentration of O_3 in the polar upper troposphere. This effect is reinforced by the reduced downwelling from the stratosphere into the troposphere of air masses characterized by low concentration CO_2 and high concentration of O_3. The bottom panel of Figure 4 shows that during the course of the stratospheric major final warming event AIRS retrieved polar CO_2 increased by ~2 ppmv while AIRS retrieved polar O_3 decreased by ~20 ppbv over five days following the event. We also investigated the impact of applying a correction for the effects of the age of stratospheric air [*Boering et al.*, 1996; *Waugh and Hall*, 2002; *Morgan et al.*, 2004] to the post-retrieved CO_2 results and found that it did not alter any of our conclusions.

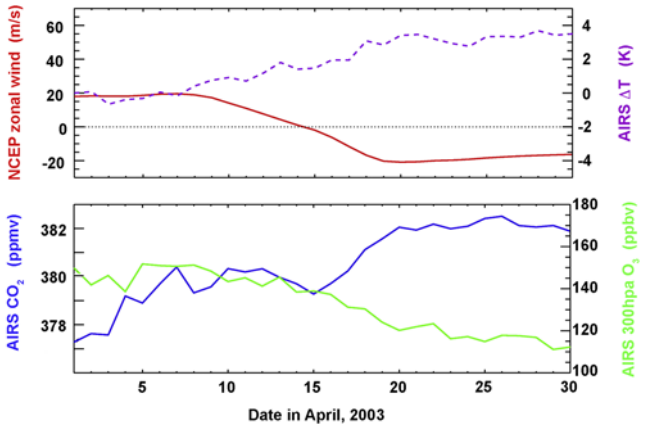

Figure 4. The major final stratospheric warming event over the northern polar region in April 2003. The top panel shows NCEP winds in m/s (red) between 60 N and 80 N at 10 hPa and AIRS temperature change (dashed purple) between 50 N and 90 N at 10 hPa. The bottom panel shows AIRS CO_2 mixing ratio in ppmv (blue) between 50 N and 90 N and AIRS O_3 mixing ration in ppbv (green) between 50 N and 90 N at 300 hPa. Data in both panels are averaged over all longitudes and the indicated latitude ranges.

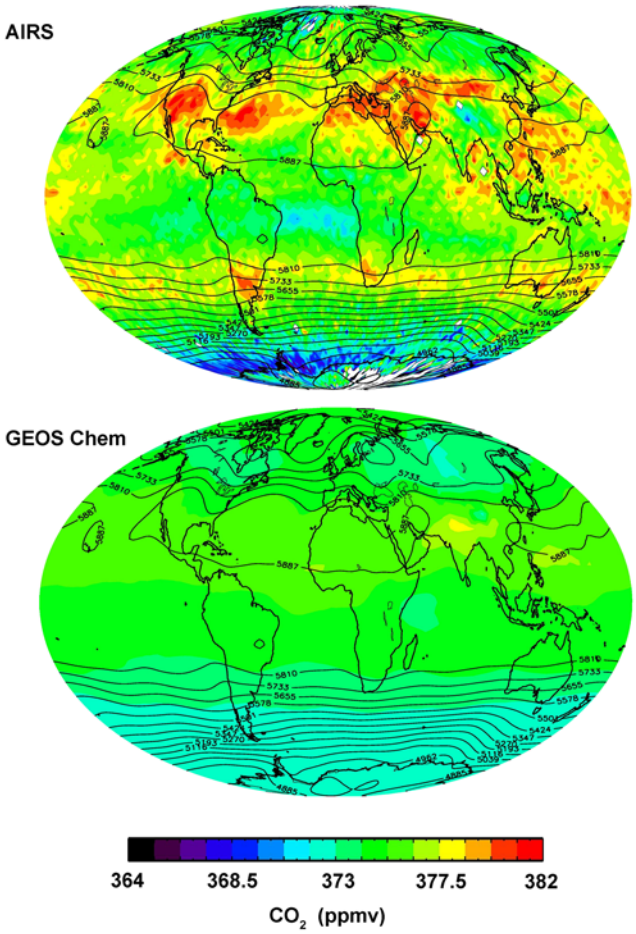

Figure 3. Comparison of geospatial distribution of retrieved and modelled mid-tropospheric CO_2 for July 2003. The top panel shows monthly mean AIRS retrieved CO_2 mixing ratios. The bottom panel shows model monthly mean GEOS-Chem Forced by Sources and Sinks CO_2 mixing ratios after convolving model profiles with the appropriate (tropical, mid-latitude or polar) weighting function of the AIRS CO_2 channels. The monthly mean 500 hPa geopotential heights from NCEP2 reanalysis are also shown in both panels. Maps of AIRS retrieved CO2 in subsequent Julys exhibit similar patterns.

4. Conclusion

[15] Our results demonstrate that satellite derived CO_2 data track weather patterns and can also be used to study the vertical and horizontal transports in the Earth's atmosphere. We have shown that CO_2 emissions by surface sources can be observed in the mid-troposphere and how they are transported around the globe. Since no model information was used to derive the distribution of CO_2, the results discussed in this paper are independent of models and thus provide an objective means to assess and improve the accuracy and performance of current three-dimensional CTMs that are used to derive the sources and sinks of CO_2.

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