

A 4-year zonal climatology of lower tropospheric CO₂ derived from ocean-only Atmospheric Infrared Sounder observations

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[1] A 4-year zonally averaged climatology of atmospheric CO₂, ocean only, between ±60° latitude has been derived from the Atmospheric Infrared Sounder (AIRS) radiances. Using only very clear fields of view, the CO₂ profile in the computed radiances is scaled until agreement is found with observations. ECMWF forecast and analysis fields are used for the temperature profile in the computed radiances. The AIRS channels used to derive CO₂ amounts are nominally sensitive to CO₂ variability in the ~300–800 mbar region (2–9 km), significantly lower in the atmosphere than that in previous studies using AIRS. Validation using aircraft measurements of CO₂ at 650 mbar indicates that the AIRS CO₂ results presented here are accurate to the 0.5–1.0 ppm level. The AIRS-derived climatology clearly exhibits the CO₂ rectifier effect, with mean CO₂ values several parts per million lower than in those in the boundary layer. The AIRS CO₂ seasonal cycle has a relatively constant amplitude of ~3 ppm from +10° to +60° latitude, which matches the boundary layer seasonal cycle amplitude near +10° latitude but is about three times smaller than that in the boundary layer amplitude at +60° latitude. Phase comparisons between the AIRS and boundary layer CO₂ seasonal cycles show the boundary layer phase leading AIRS in the Northern Hemisphere until ~+10° latitude, where the phases cross and the AIRS higher-altitude CO₂ begins to lead the boundary layer phase down to ~–10° latitude. These results may offer new insight into CO₂ interhemispherical transport. Growth rates derived from the AIRS CO₂ climatology are 2.21 ± 0.24 ppm/year, in good agreement with in situ measurements.

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1. Introduction

[2] Atmospheric CO₂ is the primary radiative forcing greenhouse gas, and its atmospheric growth rate has been rising steadily in the past few decades because of increasing global emissions [Raupach *et al.*, 2007]. Reliable estimates of climate change depend upon our ability to forecast atmospheric CO₂ concentrations, which requires knowledge of the CO₂ sources, sinks, and atmospheric transport. Inversion studies, [see, for example Denning *et al.*, 1995; Gurney *et al.*, 2003] generally use relatively sparse in situ boundary-layer CO₂ measurements, coupled with an atmospheric transport model to estimate source and sink regions and fluxes. Input data for these studies are relatively sparse, and heavily weighted to the Northern Hemisphere land sites. Constraining CO₂ sinks with existing data has been especially difficult since sinks involve large geographic areas, including the oceans. Moreover, transport of CO₂ from the boundary layer to the free troposphere is not well understood, but may be key for identification of sink regions.

[3] Two very recent studies [Stephens *et al.*, 2007; Yang *et al.*, 2007] emphasized the importance of using information on the vertical extent of CO₂ to further constrain transport and flux models. Stephens *et al.* [2007] found that only three (out of twelve) TransCom 3 transport models [Gurney *et al.*, 2003] could closely reproduce the CO₂ vertical distribution derived from a rather limited number of aircraft flights. These three particular models predicted very different flux estimates than the other nine models, strongly suggesting a weaker northern uptake of CO₂ and weaker tropical emission than previous “consensus estimates”.

[4] Yang *et al.* [2007] also found that the growing season net flux in the Northern Hemisphere is ~28% larger than predicted by models using column-averaged mixing ratios of CO₂ and partial columns derived from aircraft profiles, rather than boundary layer values. They attributed this new result to their use of the column CO₂ as the primary measurement, since it is less sensitive to vertical mixing errors in the transport models.

[5] Sufficiently accurate satellite measurements of CO₂ would greatly enhance our understanding of the global carbon cycle, by providing a much higher spatial and temporal data density. Profile information from satellite measurements may also be able to enhance the improvements discussed above by Stephens *et al.* [2007] and Yang *et al.* [2007]. Near infrared remote sensing of CO₂ can provide the

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atmospheric CO₂ column [Barkley *et al.*, 2007; Buchwitz *et al.*, 2007a, 2007b; Houweling *et al.*, 2005], while infrared sounders can potentially measure CO₂ in the free troposphere [Chédin *et al.*, 2003a, 2003b; Engelen *et al.*, 2004; Engelen and McNally, 2005; Crevoisier *et al.*, 2004; Chahine *et al.*, 2005]. Both of these measurements, separately, or in combination [Barkley *et al.*, 2006b], could potentially contribute to our understanding of the global climate budget if they had sufficient accuracy and well understood error characteristics.

1.1. This Work

[6] We present here a new 4-year zonally-averaged climatology of lower-tropospheric CO₂ derived from the Atmospheric Infrared Sounder (AIRS) flying on NASA's Aqua satellite. This climatology is restricted to clear, ocean-only observations in the $\pm 60^\circ$ latitude range. What distinguishes this work from previous AIRS retrievals of CO₂ is our use of channels that peak at ~ 550 mbar, rather than near ~ 200 mbar, with a continuous 4-year climatology that allows accurate measurements of growth rates and seasonal variability as a function of latitude.

[7] Our primary measurement uses only a single AIRS channel sensitive to CO₂, centered at 791.7 cm^{-1} , because of its combined low sensitivity to temperature and high sensitivity to CO₂. We distinguish between temperature and CO₂ variations by using European Center for Medium-range Weather Forecasts (ECMWF) analysis/forecast model data for the atmospheric temperature profile. This overcomes one of the major difficulties in AIRS-only retrievals of CO₂, but, of course, makes our results dependent on the accuracy of the ECMWF fields. We perform similar retrievals of CO₂ with AIRS using channels in the 2400 cm^{-1} region that are more sensitive to temperature than the 791.7 cm^{-1} channel, and use the difference in the CO₂ product from these two spectral regions as a diagnostic on the accuracy of the ECMWF temperature fields. In this paper we will often refer to the 791 cm^{-1} region as long-wave, LW, and the 2400 cm^{-1} region as short-wave, SW.

[8] The CO₂ variability reported here represents very small signatures in the AIRS radiances. The brightness temperature of the more sensitive 791.7 cm^{-1} channel only decreases by approximately -0.03 K for a 1 ppm positive offset of the CO₂ profile. The 2400 cm^{-1} channels are even less sensitive to CO₂. The full range of CO₂ values derived from the AIRS spectra for the four years examined here is 18 ppm, which only represents a brightness temperature range of 0.54 K . The results presented here therefore highlight the extreme stability of both the AIRS instrument and the ECMWF model fields.

[9] A major advantage of our simple approach using clear-only fields of view is the ability to easily process, and re-process, this 4-year data set. Partial validation of our climatology is made by comparison to the NOAA GLOBALVIEW [GLOBALVIEW-CO₂, 2006] aircraft flights over ocean, and the GLOBALVIEW Marine Boundary Layer (MBL) CO₂ product, which is a smoothed zonal representation of a large number of globally distributed in situ measurements and has an estimated accuracy of ~ 0.3 ppm. The comparisons to MBL help establish the plausibility of our results and stimulate further investigations. In this work we introduce our retrieval methods and

validation, comparisons to global CO₂ models will be presented in a forthcoming paper.

1.2. Previous Work

[10] Infrared sensing of CO₂ began with the pioneering work of Chédin *et al.* [2002, 2003b] using the NOAA/TOVS infrared sounder. The more recent work of Chédin *et al.* [2003b] used a neural network retrieval approach that could differentiate between temperature and CO₂ by including Microwave Sounding Unit (MSU) radiances in the process, since MSU radiance are independent of CO₂ but not temperature. Their infrared CO₂ Jacobians peaked at ~ 300 mbar, and they analyzed 4 years covering the 1987–1991 time frame between $\pm 20^\circ$ latitude. Their measurement standard deviations were about 3 ppm. They also successfully retrieved mean CO₂ growth rates over the $\pm 20^\circ$ latitude range of ~ 1.75 ppm/year that agreed quite well with in situ measurements.

[11] The SCIAMACHY instrument on ENVISAT allows the measurement of the CO₂ column, mostly over land, with a precision that gives agreement with models to within ~ 2 ppm for the amplitude of Northern Hemisphere seasonal cycles. The SCIAMACHY measurements may have biases of 4% or less [Barkley *et al.*, 2007], which are difficult to assess because of the relatively high variability of CO₂ over land sites and the lack of in situ data. Recently Buchwitz *et al.* [2007a, 2007b] measured the CO₂ rate of growth for the 2003–2006 time period with SCIAMACHY, and found agreement with the NOAA CarbonTracker [Peters *et al.*, 2007] in the 1 ppm/year range. At present, it is not clear if the SCIAMACHY data is sufficiently unbiased, or has well enough characterized errors, to be used for inverse modeling of CO₂ [Barkley *et al.*, 2006a; Tiwari *et al.*, 2006].

[12] AIRS (Atmospheric Infrared Sounder) [Aumann *et al.*, 2003] on NASA's Aqua satellite platform provides a wealth of high-spectral resolution channels with sensitivity to CO₂, but with limited boundary-layer information, unlike SCIAMACHY. However, AIRS does have the ability to potentially provide several independent pieces of information on the CO₂ profile above the boundary layer.

[13] A number of previous studies have used AIRS channels peaking in the upper troposphere (200 mbar range) to retrieve CO₂ [Engelen *et al.*, 2004; Engelen and McNally, 2005; Crevoisier *et al.*, 2004; Chahine *et al.*, 2005]. These nominal 200 mbar measurements make them significantly less sensitive to the immediate effects of CO₂ sources and sinks. They are more dependent on transport, and could possibly be used to constrain transport model biases, although Chevallier *et al.* [2005] suggest that the AIRS CO₂ biases must be kept to a few tenths of a ppm for assimilation into a transport model.

[14] The most comprehensive AIRS CO₂ product by Engelen and McNally [2005] covers a period of 15 months. They quote monthly *random* mean errors of 1–6 ppm (1–3 ppm for the latitudes reported in this paper). Tiwari *et al.* [2006] have made detailed comparisons between a model and their retrievals with qualitatively good agreement, especially for amplitude of the seasonal cycles. However, the transport model shows significant growth of CO₂ in the Northern Hemisphere winter that is absent in their AIRS CO₂ weighted column [see Tiwari *et al.*, 2006, Figure 8].

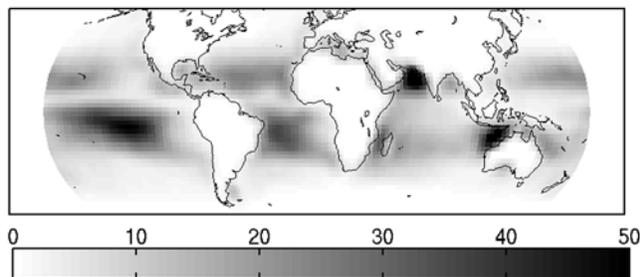


Figure 1. Density plot (in units of 1000) of clear AIRS FOVs used to derive the AIRS CO₂.

Our lower altitude CO₂ product reported here does indeed show this Northern Hemisphere winter growth of CO₂.

[15] *Chahine et al.* [2005] introduced an independent CO₂ retrieval using AIRS that also uses channels that peak in the ~ 300 mbar region. He reports results for two years of AIRS match ups with the JAL airline measurements [*Matsueda et al.*, 2002], with a monthly mean standard deviation of 1.2 ppm.

[16] The neural-network approach of *Chédin et al.* [2003a] has also been applied to AIRS by *Crevoisier et al.* [2004] using channels with the CO₂ Jacobian peaking at ~ 200 mbar. They analyzed seven months of data in the $\pm 20^\circ$ latitude range with an estimated precision of 2.1–2.5 ppm for monthly means with $15^\circ \times 15^\circ$ spatial averaging. They indicate low biases, ~ 0.9 ppm, relative to the work of *Matsueda et al.* [2002] (JAL using GLOBALVIEW terminology).

[17] Finally, *Aumann et al.* [2005] tracked two AIRS channels over time between $\pm 30^\circ$ latitude. One channel is mainly sensitive to N₂O only, while the other channel is dominated by CO₂. He showed that if you assume an N₂O growth rate, that the relative change in radiance in these two channels lead to a CO₂ growth rate that agrees very well with in situ measurements.

[18] In summary, although there have been a number of satellite measurements of atmospheric CO₂, they have either probed the middle to upper troposphere (200–300 mbar peak sensitivities) or have measured the atmospheric CO₂ column (using near-infrared sensors which work primarily over land). The results presented here, although only over ocean, are the first to use thermal infrared satellite data to measure CO₂ in the lower-troposphere. In addition, our CO₂ retrievals cover a much longer time span, with higher accuracy, than previous infrared remote sensing studies. This comprehensive ocean climatology allows detailed analysis of the latitude dependence of the lower-tropospheric CO₂ seasonal cycle amplitude and phase, and of CO₂ growth rates.

2. AIRS Satellite Data

[19] AIRS measures 2378 high-spectral resolution infrared radiances between 650 and 2665 cm⁻¹ with a nominal resolving power ($\lambda/\Delta\lambda$) of 1200 and nadir footprint of 13 km. Flying on board NASA's EOS Aqua spacecraft, AIRS has been operational since September 2002, supplying almost continuous global measurements twice each day in a sun-synchronous orbit with very stable equator crossing times of 1:30 am and 1:30 pm, and 705 km orbital altitude.

AIRS primary purpose was to improve operational weather forecasting [*Chahine et al.*, 2006], but the excellent stability of AIRS is now allowing studies related to climate and monitoring of atmospheric composition.

[20] The CO₂ climatology reported here is derived from AIRS night-only, clear fields of view (FOVs), over ocean. The algorithm for selecting clear FOVs has been previously described by *Strow et al.* [2006], and is primarily based on a uniformity filter and several spectral tests. This filter is very similar to one described by *Aumann et al.* [2006], and returns only about 1% of all ocean observations. Following *Aumann et al.* [2006], we estimate that our clear FOVs have a residual amount of cloud contamination that produces a nominal 0.2 K cold bias in the spectra.

[21] A density map of the FOVs returned by the clear detection algorithm shown in Figure 1 indicates that our individual samples are widely dispersed, although some locations are more intensively sampled because of prevailing clear conditions. Obviously, the reduction of statistical errors by zonal averaging may introduce errors that can complicate the interpretation of the CO₂ climatology. This may be especially true for the Indian/Arabian Ocean where the Asian continent occupies the northern latitudes, giving more of a continental rather than ocean flavor to the CO₂ climatology.

[22] A main goal of this work is to derive CO₂ concentrations at lower altitudes than previous studies. The existing retrievals of CO₂ with AIRS used channels peaking in the 200–300 mbar region because the authors wished to avoid channels contaminated by either other trace gases or by surface emission. A comprehensive examination of contamination of AIRS channels has been performed in the work of *Crevoisier et al.* [2003] and *Chédin et al.* [2003a]. For example, Figure 6 of this last reference shows that many of the mid-tropospheric AIRS channels in the 700–780 cm⁻¹ region have significant O₃ contamination. Many CO₂ channels have significant surface contamination that other investigators wished to avoid. *Crevoisier et al.* [2003] suggest a list of candidate channels for CO₂ retrievals in the wave number ranges from 664–741 cm⁻¹ and 2249–2391 cm⁻¹, most of which have CO₂ Jacobians that peak far higher than the 791.7 cm⁻¹ channel we use. The only low-peaking channels they selected are the 2388–2391 cm⁻¹ SW channels. However, note that at present, actual CO₂ retrieval results have only been reported for channels peaking in the 200–300 mbar range.

[23] Since we use ECMWF for the temperature profile, we wish to minimize the effect of errors in the temperature profile. A useful metric for channel selections is the ratio

$$\frac{dBT/dCO_2}{dB T/dT} \quad (1)$$

where dT represents a uniform (1 K) offset to the air temperature profile and dCO_2 is a uniform (-10%) offset to the CO₂ profile. Here we wish to maximize sensitivity to CO₂ while minimizing sensitivity to temperature profiles errors. Figure 2 plots this ratio for the 4 and 15 μm regions sensitive to CO₂. The AIRS channel centered at 791.7 cm⁻¹ easily has the largest sensitivity ratio. Furthermore, Figure 3 shows that the CO₂ Jacobian for this channel peaks at ~ 500 – 550 mbar or ~ 5 km, rather than at 200–300 mbar,

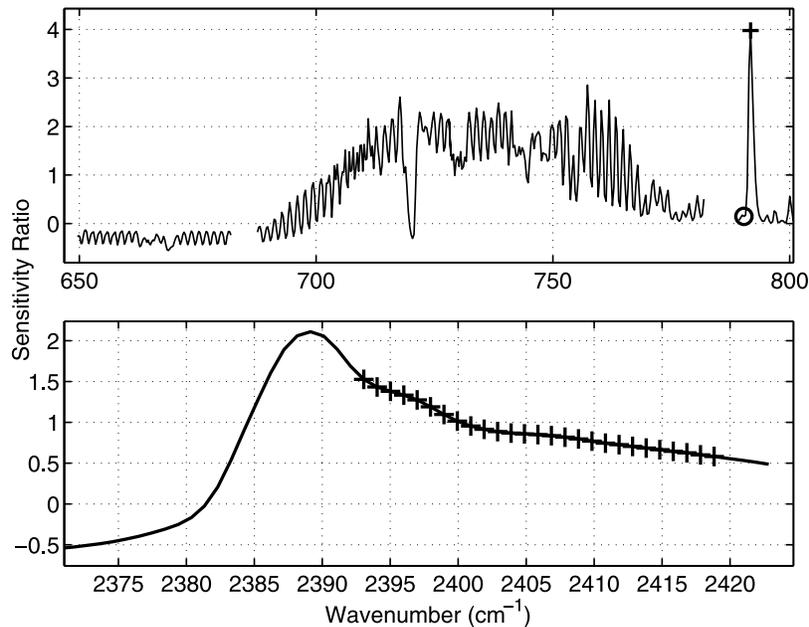


Figure 2. Ratios of CO₂ to profile temperature sensitivity in observed $B(T)$ s or $(dB T/dCO_2)/(dB T/dT)$. T is the atmospheric temperature profile, which was shifted by a uniform 1 K offset. The CO₂ profile was decreased with a uniform -10% offset. In the top, the 791.7 cm⁻¹ CO₂ channel is denoted with a plus, and the 790.3 cm⁻¹ channel is denoted with a circle. The SW channels used to derive CO₂ are denoted with plus symbols in the bottom.

or 9–12 km, used in previous studies of CO₂ with AIRS [Engelen *et al.*, 2004; Engelen and McNally, 2005; Crevoisier *et al.*, 2004; Chahine *et al.*, 2005]. The 4 μm channels have a lower sensitivity ratio, around unity, partly because they are more temperature sensitive and partly because they are weaker transitions. However, as seen in Figure 3 the CO₂ Jacobians for these channels are quite similar to the 791.7 cm⁻¹ CO₂ Jacobian, and probe the mid-

to lower-troposphere. The channels used for our SW CO₂ retrieval stop at 2393 cm⁻¹ because stratospheric sensitivity grows quickly as you move to lower wave numbers.

[24] Both the 791.7 cm⁻¹ and 2400 cm⁻¹ CO₂ channels will have significant surface contamination that must be properly modeled so it is not confused with CO₂. For the 791.7 cm⁻¹ transition, a very close channel at 790.3 cm⁻¹ has no CO₂ absorption and can be used to derive the

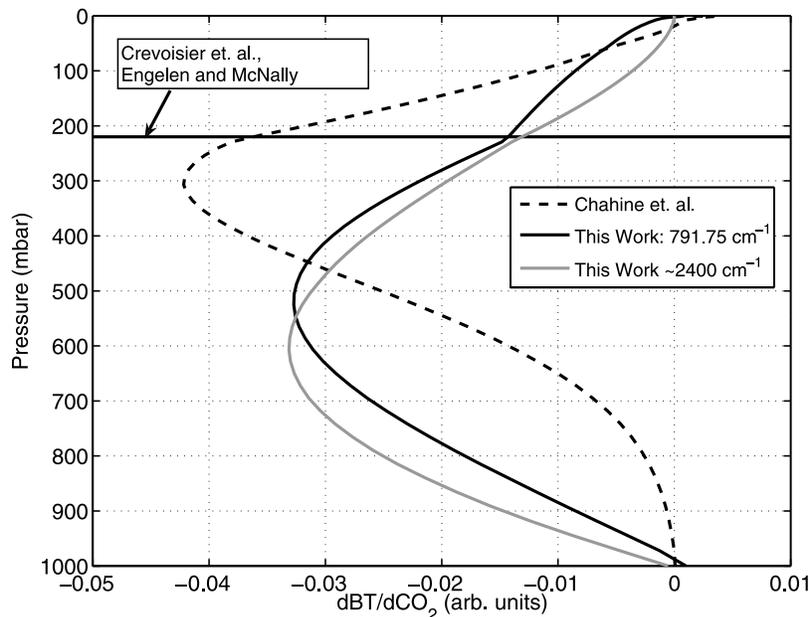


Figure 3. CO₂ Jacobians $(dB T/dCO_2)$ for the channels used in this work and for the work of Chahine *et al.* [2005]. Also shown are the reported peak locations of the CO₂ Jacobians used by Crevoisier *et al.* [2004] and Engelen and McNally [2005].

effective surface emission for the 791.7 cm⁻¹ channel. In the 2400 cm⁻¹ region the CO₂ transmittance drops rapidly (but not to zero) past 2400 cm⁻¹, allowing a simultaneous retrieval of the effective surface emission and CO₂ column offset.

[25] Although the Masuda surface emissivity is quite accurate, the very stringent accuracy requirements for the CO₂ retrieval led us to re-estimate the effective surface temperature (which nominally includes the surface emissivity) close to the CO₂ channels. In addition, small residual effects of un-detected low clouds, and possibly improperly modeled water vapor continuum, may be present in our computed radiances. Since our 790.3 cm⁻¹ surface channel is only 1.4 cm⁻¹ away from the 791.7 cm⁻¹ CO₂ channel, we assume these surface emissions are identical for these two channels. Similarly for the 2400 cm⁻¹ region, we assume surface emissions are identical in the 2393–2418 cm⁻¹ range used for the SW CO₂ retrieval.

[26] A major assumption of this work is that the ECMWF temperature fields are statistically accurate enough to allow meaningful CO₂ retrievals. We partially test this assumption by comparing the LW to SW CO₂ (see section 3.2). ECMWF uses radiosonde measurements as the “anchoring network” of observations for the ECMWF tropospheric temperatures [Auligne *et al.*, 2007], with no bias correction. This is in contrast to infrared satellite data assimilated by ECMWF (such as AIRS), which is bias-corrected to provide better agreement with the model and radiosondes. In large part the infrared satellite data is used to lower the standard deviation of the model rather than to provide model offsets.

[27] We also note that comparisons between ECMWF computed and observed AIRS radiances (for CO₂ dominant channels) generally have standard deviations that are almost equivalent to the AIRS instrument noise, indicating that ECMWF is tracking the true atmosphere temperatures very closely.

3. CO₂ Retrieval Method

[28] The starting point for our CO₂ retrieval are the nominally clear AIRS radiance observations, and the nearest matching ECMWF operational analysis or forecast model profiles. ECMWF provides an analysis at 00, 06, 12, and 18 UTC, 3-hour forecasts at 03, and 15 UTC, and 9 hour forecasts at 09 and 21 UTC. Prior to February 2006 these fields were on a T511L60 grid (0.5° lat/lon resolution, 60 vertical layers), which ECMWF subsequently upgraded to a T799L91 grid (0.25° lat/lon, 91 vertical layers).

[29] We first compute synthetic radiances using the ECMWF profile and sea-surface temperature, the Masuda sea surface emissivity model [Masuda *et al.*, 1988], and a default CO₂ amount of 370 ppm. The radiative transfer model used for these calculations is a stand-alone version of the AIRS operational radiative transfer algorithm, SARTA [Strow *et al.*, 2003, 2006]. In order to reduce bias errors due an inaccurate ECMWF SST or total column water (TCW), we first derive these quantities from the observed spectra. The fitted SST differs from the ECMWF SST because of the effects of evaporative cooling of the sea surface, and because of small amounts of residual cloud contamination.

[30] The SST and TCW fits use a combination of windows channels, and channels containing weak water vapor lines, all in the 2600 cm⁻¹ region, as previously described [Strow *et al.*, 2006]. A number of window channels in this region contain only ~0.3 K of atmospheric absorption, and are therefore ideal for deriving an effective SST. We also simultaneously minimize the bias for several weak water line channels, giving us an improved TCW. The TCW is modified by scaling the ECMWF water vapor profile with a constant multiplier. A re-calculation of synthetic AIRS spectra using these two new parameters lowers the standard deviation of the biases in the LW (10–12 μm) by a factor of three. Our mean (±45° latitude) TCW agrees with the ECMWF model TCW to ~3%.

[31] The CO₂ amount is retrieved by a linear least-squares fit of the following set of equations that relate the bias to the derivatives of the brightness temperature for channel *i*, the CO₂ column scaling factor, δCO₂, and effective surface temperature offset δT_s,

$$BT_i^{obs} - BT_i^{cal}(\text{ECMWF}) = \frac{dB_i}{dCO_2} \delta CO_2 + \frac{dB_i}{dT_s} \delta T_s. \quad (2)$$

For the LW CO₂ climatology we use only two channels, while for the SW climatology we use 26 channels. The LW and SW climatologies are completely separate products, each derived from simultaneous fits of the above equation to only LW channels, or, to only SW channels. Validation and detailed analysis presented here uses only the LW CO₂ climatology, which we consider more accurate.

[32] All computed brightness temperatures, and their surface and CO₂ derivatives were computed separately for every observed FOV. The above set of simultaneous equations were originally solved for the CO₂ amount for each FOV. This is time-consuming procedure, given the large number of FOVs. Instead, we aggregated these data into 1-day, 4° latitude bins before solving for CO₂, which gives essentially identical results.

[33] The highest two latitude time series can contain significant numbers of missing points because of lack of clear FOVs. Figure 4 plots three individual LW time series of observed BTs that illustrate the significant number of missing points at the higher latitudes (54°S is shown). Also shown in the bottom of Figure 4 is the mean Northern Hemisphere BT bias, showing how the observed BTs are getting colder with time, modulated by the seasonal cycle, because of increasing CO₂. The gray curve in the bottom is the associated mean times series of dT_s, which does not exhibit any long-term trend.

[34] The final CO₂ product has dimensions of 30 latitude bins (–60 to +60° latitude in 4° steps) and 1460 days (4 years). Most results shown in this paper use the LW AIRS CO₂ product, which we consider to be more accurate than the SW AIRS CO₂ product. The SW CO₂ product is, however, used for diagnosing errors, especially relative to ECMWF temperature profiles.

3.1. Calibration

[35] The accuracy of a computed radiance is often limited by the accuracy of the molecular spectroscopy used to compute the atmospheric transmission and emission terms in the radiative transfer equation. It is very difficult to get

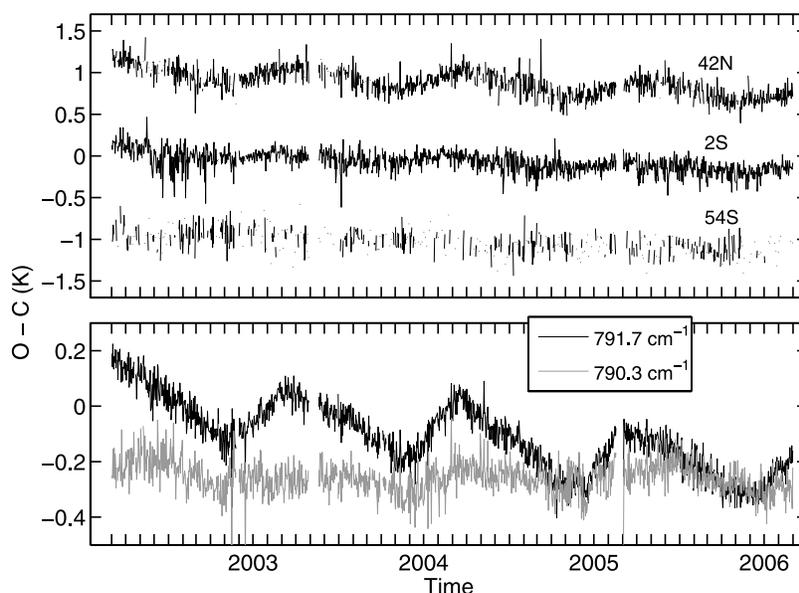


Figure 4. (Top) Examples of 4° latitude binned biases between observed and computed 791.7 cm⁻¹ brightness temperatures. The 42N/54S biases have been offset by +1K/-K respectively. (Bottom) Northern Hemisphere mean biases for both the LW CO₂ channel (791.7 cm⁻¹) and the surface channel (790.3 cm⁻¹). Note that, as expected, there is no long-term trend in the surface channel bias. For this plot, and all subsequent plots versus time, tick marks denote the first day of each month, and the year labels are centered below the 1 July tick marks.

agreement among laboratory measurements of molecular vibration-rotation transition strengths, widths, and shapes to even the 2% level. If one takes 2% as the accuracy of the CO₂ optical depths used in the radiative transfer, this translates, for the 791 cm⁻¹ CO₂ transition, to an error of 0.22 K in brightness temperature, or an error of about 8 ppm in the CO₂ volume mixing ratio. Given that we expect the AIRS stability to be <0.016 K/year [Aumann *et al.*, 2006], or equivalently <0.5 ppm of CO₂ per year, our measurements will be far more precise than accurate.

[36] The CO₂ record at Mauna Loa provides a very convenient one-time, one-value offset calibration for the AIRS CO₂ measurements. The Mauna Loa CO₂ accuracy is well-established (at the sub-ppm level) and is commonly used as a baseline for comparisons among other measurements. Mauna Loa is particularly suited for the AIRS CO₂ calibration since it is located at ~650 mbar, close to the nominal 550 mbar peak of our CO₂ sensitivity function. In addition, at the latitude of Mauna Loa (19.5°N) atmospheric CO₂ is relatively well mixed, especially compared to other latitudes. We therefore offset our 4-year record of CO₂ to agree with the mean of the Mauna Loa CO₂ values, using our zonal CO₂ values within 8° latitude of Mauna Loa, and only observations in the Pacific Ocean.

[37] This calibration may take into account factors other than CO₂ spectroscopy errors, which could include AIRS radiometric calibration errors and errors in our radiative transfer model separate from transmittance errors. In addition, this one-time calibration adjusts our vertically-averaged CO₂ amount into a nominal 650 mbar value. Between ±60° latitude the peak of the 791 cm⁻¹ CO₂ Jacobian only varies by about 50 mbar. The 2400 cm⁻¹ kernel function peaks at a slightly lower altitude than the 791 cm⁻¹ channel, around 600 mbar.

[38] This calibration led us to increase the AIRS-derived CO₂ amounts by +7.2 ppm for the 791.7 cm⁻¹ transition and +9.5 ppm for the 2400 cm⁻¹ SW region CO₂ measurement. These values are well within expected spectroscopic and radiometric error budgets. For example, the HITRAN database [Rothman *et al.*, 2005] uncertainty estimate for the 791.7 cm⁻¹ CO₂ Q-branch line strengths is 2–5%, which translates into an uncertainty in the CO₂ amount of 8–18 ppm. Note that in the 2400 cm⁻¹ region the AIRS radiative transfer model transmittances were previously adjusted, on the basis of coincident radiosonde measurements [see Figure 9 of Strow *et al.*, 2006], and therefore the 9.5 ppm calibration offset derived for the SW CO₂ retrieval cannot be interpreted as the true spectroscopy error.

[39] The AIRS pre-launch radiometric calibration uncertainty was ~0.1 K near 790 cm⁻¹ [Pagano *et al.*, 2003], which has been validated in orbit to less than 0.2 K [Aumann *et al.*, 2006]. The 791.7 cm⁻¹ channel sensitivity to CO₂ is 0.03 K/ppm, which translates to a minimum uncertainty in CO₂ because of radiometric error of ~3–7 ppm (for a 0.1–0.2 K radiometric uncertainty) that may occur in addition to spectroscopic errors.

[40] All of these considerations suggest that these small calibration adjustments of +7.2 ppm for the LW, and +9.5 ppm for the SW are well within the bounds of our estimated absolute errors.

[41] Figure 5 shows our smoothed zonally averaged CO₂ values for the first four years of AIRS operations averaged over the 19.5 ± 4° latitude range, with and without the +7.2 ppm calibration offset. The bottom shows the difference between the Mauna Loa and AIRS CO₂ as a function of time. Note that the AIRS CO₂ is zonally averaged over the Pacific Ocean while the Mauna

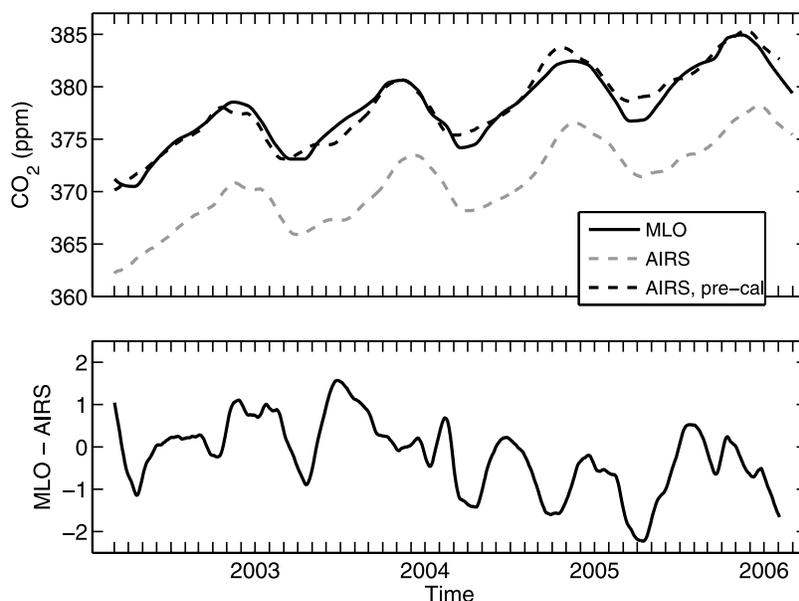


Figure 5. (Top) The NOAA-ESRL GLOBALVIEW CO₂ record at Mauna Loa and the AIRS (LW) derived CO₂ record (Pacific Ocean only) before and after calibration adjustment to the Mauna Loa amount. (Bottom) Mauna Loa minus AIRS CO₂ (after CO₂ offset adjustment).

Loa data is a point measurement, so some disagreement is expected because of the zonal variability observed by AIRS.

[42] For plotting purposes, both in this figure and subsequent figures, we smooth and fill missing data in the CO₂ time series using the Singular Spectrum Analysis (SSA) approach [Ghil *et al.*, 2002; Kondrashov and Ghil, 2006]. Only the ± 56 and $\pm 60^\circ$ latitude CO₂ time series needed significant filling. A correlation length of 1 year was used to fill missing points [Kondrashov and Ghil, 2006]. For smoothing, we used a correlation length of 40 days, which does not necessarily remove behavior over a shorter time period. We do not use these smoothed results in our main analysis of the CO₂ time series (fits to equation (3)), but instead fit to the original un-smoothed, un-filled CO₂ time series.

[43] The gray dashed curve in the top of Figure 5 is the raw AIRS CO₂ at the Mauna Loa site. A least-squares fit of both of these time series (see section 5.3 for details) indicates that the Mauna Loa seasonal cycle is leading the AIRS seasonal cycle by 28 days. We increased the AIRS phase by this amount, and added in the 7.2 ppm calibration offset, before computing the Mauna Loa minus AIRS curve shown in the bottom.

[44] The Mauna Loa minus AIRS time series has repeated minima in the fall of each year, approximately 3–4 weeks after the Northern Hemisphere boundary layer CO₂ begins rising after the summer draw-down. AIRS may be detecting slightly higher CO₂ amounts than Mauna Loa, since the AIRS zonal average contains scenes closer to continents. The additional minimum in MLO minus AIRS in late winter of 2005 may be related to the slow growth in the boundary layer compared to the AIRS measurements that are discussed in more detail in section 5.3 (see Figure 21).

[45] Since the peak disagreements between Mauna Loa and AIRS are generally <1 ppm in Figure 5, we estimate the accuracy of this calibration to be in the 1 ppm range. A

more detailed analysis of the accuracy of our derived CO₂ product, using other data sources, is given in section 4.

3.2. LW Versus SW CO₂

[46] Comparisons between our AIRS LW and SW CO₂ climatology provides some degree of validation, since these two spectral regions are far apart and are subject to different kinds of interfering processes. For example, the 791 cm⁻¹ brightness temperatures can be depressed below the sea surface temperature by up to 7 K in the tropics because of the water vapor continuum, while the max depression in the SW channels due to water vapor is below 1 K. The channel used to derive the surface temperature \times emissivity product in the LW has effectively no CO₂ absorption. In the SW all channels used to determine the surface emission have significant CO₂ absorption. Although the 791.7 cm⁻¹ channel is less temperature sensitive than the SW channels, it is potentially more sensitive to cloud contamination because of the wavelength sensitivity of the Planck function.

[47] The differential sensitivity of the SW and LW CO₂ to ECMWF model fields is illustrated in Figure 6, where we plot our mean Northern Hemisphere CO₂ climatology derived from both channels sets. We removed a slight bias of 0.25 K between the SW and LW CO₂ for the time period up to 1 February 2006 in order to highlight the change in the CO₂ values after this time. The bottom of this plot shows the smoothed difference between these two time-series, which clearly highlights a sharp change of ~ 2 –3 ppm in February 2006 that we trace to changes in the ECMWF model (see http://www.ecmwf.int/products/data/operational_system/evolution/evolution_2006.html#1February2006). The sensitivity calculations plotted in Figure 2 are for a U.S. standard atmosphere and show that the LW CO₂ retrieval is about four times less sensitive to ECMWF than the SW results. More detailed calculations show that this ratio is ~ 2.6 at the equator and grows to ~ 4.9 at the higher latitudes.

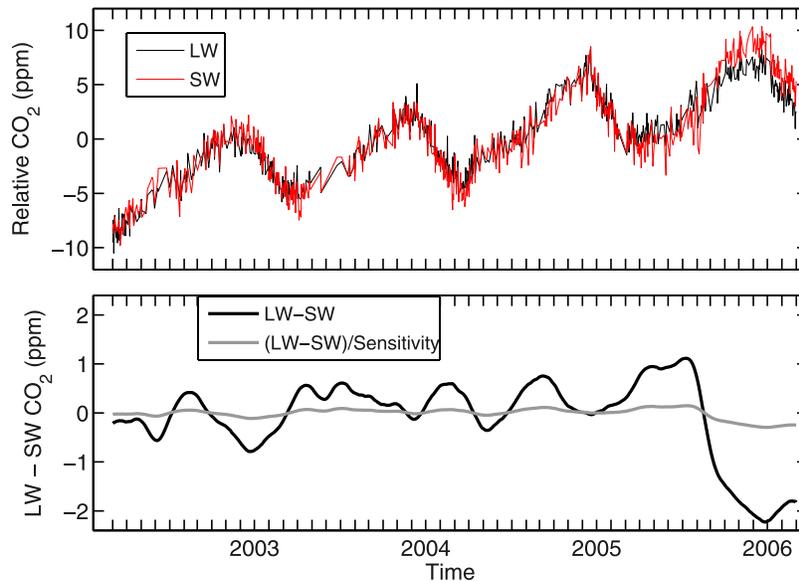


Figure 6. (Top) LW and SW AIRS CO₂, 5° to 45° latitude mean. (Bottom) Smoothed LW minus SW CO₂; the same quantity was divided by the differential sensitivity between LW and SW to temperature profile errors. (The differential sensitivity is the ratio of the channel average of equation (1) evaluated for the SW divided by equation (1) evaluated for the LW CO₂ channel.) The gray curve is an estimate of the uncertainty in our LW AIRS CO₂.

[48] One way to estimate of the effects of ECMWF model error on our LW CO₂ product is to divide the difference between the LW and SW CO₂ by this sensitivity ratio, which is the gray curve in the bottom of Figure 6. While this is probably an underestimate of the impact of ECMWF errors on the LW CO₂, it does suggest that they may be less than 1 ppm. If the ECMWF temperature shift is distributed non-uniformly in the vertical, this error may be larger, since

our sensitivity calculations used a vertically constant temperature offset.

[49] A comparison of the 3-year mean of the LW and SW CO₂ versus latitude is shown in Figure 7. The SW CO₂ has been normalized to give the same mean CO₂ as the LW, averaged over latitude. These two curves are similar but do exhibit systematic differences of up to 1 ppm, depending upon latitude. These may occur because the SW CO₂

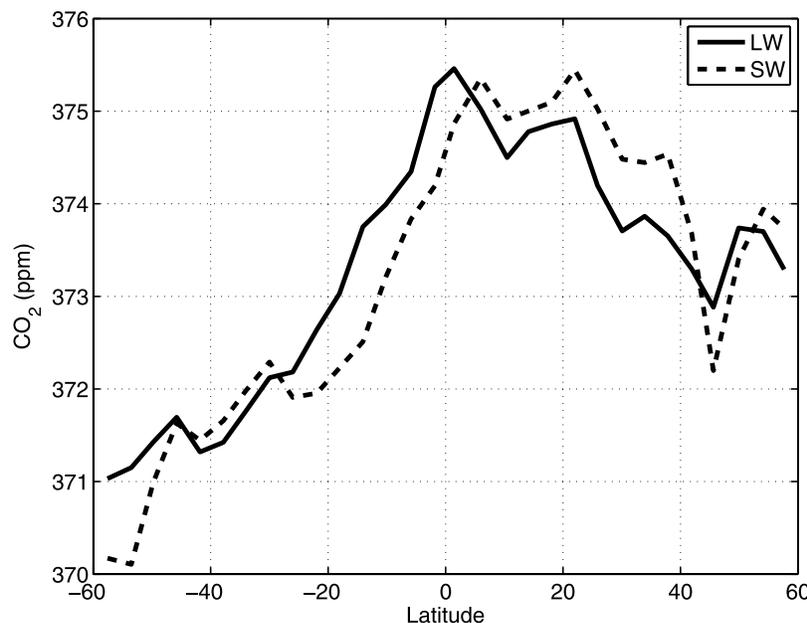


Figure 7. Three-year mean (2003–2005) of detrended AIRS CO₂. The solid line is the results reported here using the LW 791.7 cm⁻¹ channel; the dashed line is the AIRS CO₂ derived from the SW 2400 cm⁻¹ region, normalized to give the same mean CO₂ as the 791.7 cm⁻¹ channel result when averaged over latitude. The error in the LW CO₂ caused by inaccuracies in the ECMWF temperature profiles should be several times smaller than the differences between the LW and SW CO₂ amounts shown here.

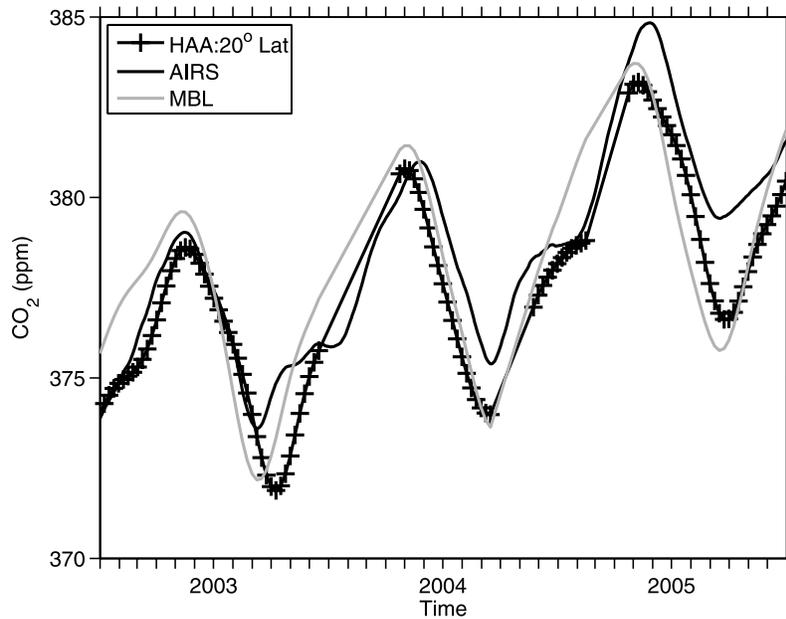


Figure 8. Molokai Island, Hawaii (HAA) CO₂ from aircraft flights at 3.5 km (650 mbar) compared with AIRS (LW) and NOAA-ESRL MBL boundary layer CO₂.

Jacobian is larger at lower altitudes and thus more sensitive to ECMWF temperature profile errors. If there is indeed more CO₂ at higher altitudes in the southern tropics, this would result in a lower SW CO₂ in that region. Conversely, above ~10° latitude, the mean CO₂ is higher at low altitudes, making the SW CO₂ somewhat higher than the LW CO₂.

4. Validation

[50] We compare the AIRS zonal CO₂ to the three aircraft flight series in the NOAA/ESRL GLOBALVIEW database that are over open ocean in the ±60° latitude range. For

two of these flight series, HAA (Molokai Island, Hawaii, +21.2° latitude, -159° longitude) and RTA (Rarotonga, Cook Islands, -21.25° latitude, -160° longitude), we compare to data taken at flight altitudes of 3.5 km (~650 mbar), close to the altitude of Mauna Loa, which we used for our calibration constant. In addition, we also compare our results to the extensive set of aircraft measurements by *Matsueda et al.* [2002] taken on commercial Japan airlines flights (JAL), at an altitude of 10.5 km (~250 mbar). The JAL CO₂ measurements are much higher in altitude than the peak of our CO₂ measurement Jacobian, although we still have significant contributions from that altitude (see

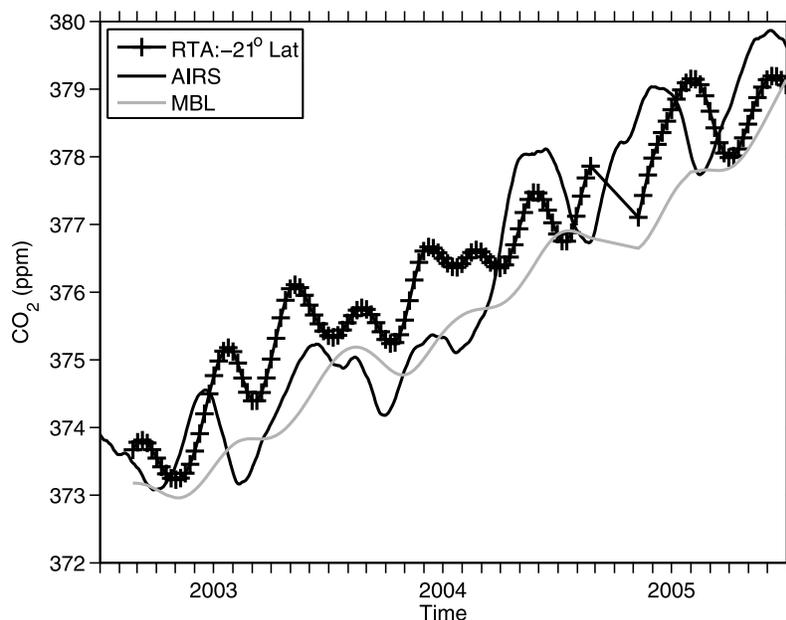


Figure 9. Rarotonga, Cook Islands (RTA) CO₂ from aircraft flights at 3.5 km (650 mbar) compared with AIRS (LW) and NOAA-ESRL MBL boundary layer CO₂.

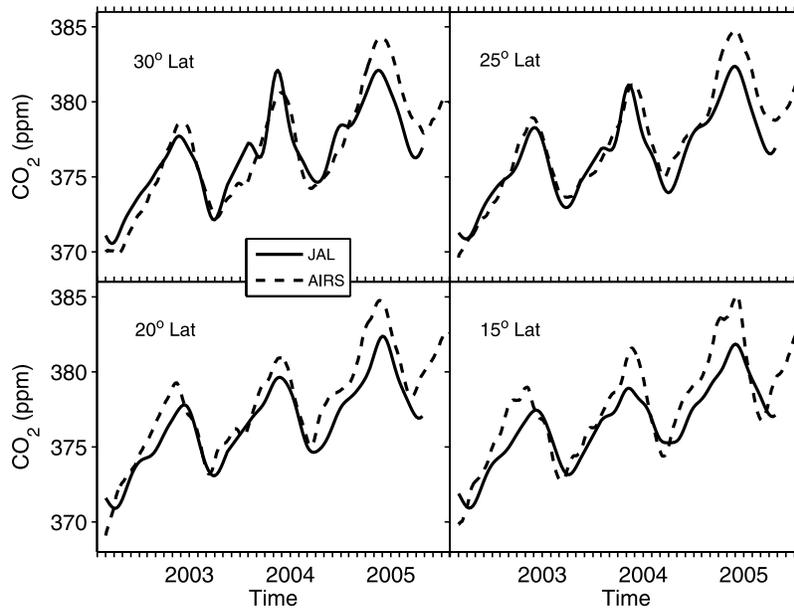


Figure 10. Japan Airlines (JAL) CO₂ measured at 10.5 km (250 mbar) compared with AIRS (LW) for 15° to 30° latitude in 5° increments.

Figure 3). For the lower altitude HAA and RTA data we also plot the NOAA GLOBALVIEW MBL values for reference.

[51] Figure 8 shows that the AIRS, HAA, and MBL time series have considerable overlap over the three years shown. Not surprisingly, HAA versus AIRS exhibits similarities to the comparison of Mauna Loa versus AIRS. For example, the AIRS zonal average is again slightly higher than HAA 3–4 weeks after the Northern Hemisphere boundary layer CO₂ begins rising after the summer draw-down. (Note that this feature cannot be seen in 2004 since HAA had a data gap during that fall.) This feature may be due to the influence of the Indian/Arabian ocean CO₂ on the AIRS

zonal CO₂ measurement (see section 5 for a more detailed discussion).

[52] HAA minus AIRS is -0.71 ppm on average, with a standard deviation of 0.86 ppm. These statistical comparisons combine the AIRS CO₂ uncertainty and the zonal variability that is folded into the AIRS CO₂ product. Generally the MBL ramp-up of CO₂ in the winter is stronger than either the HAA or AIRS behavior (presumably because of decreased convection to higher altitudes) while both HAA and AIRS CO₂ values follow the draw-down of CO₂ more closely during the middle of the year (increased convection).

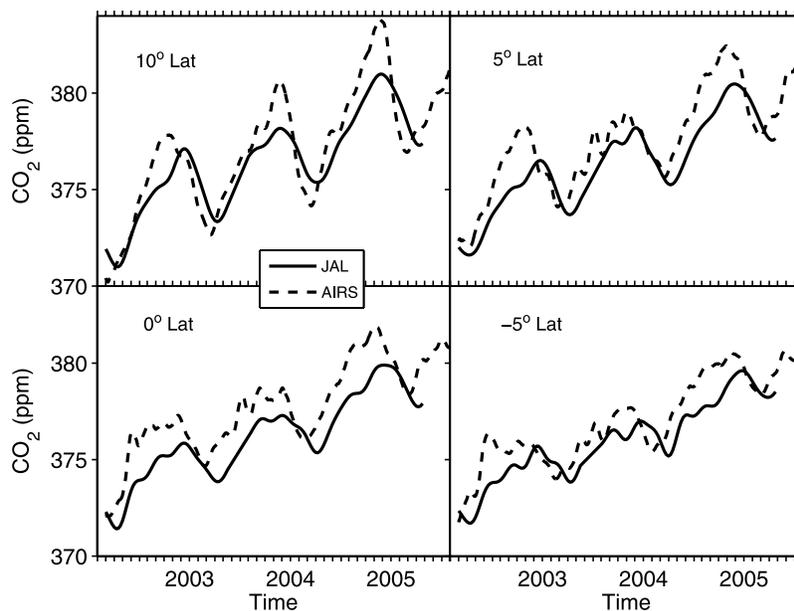


Figure 11. JAL CO₂ measured at 10.5 km (250 mbar) compared with AIRS (LW) for -5° to $+10^{\circ}$ latitude in 5° increments.

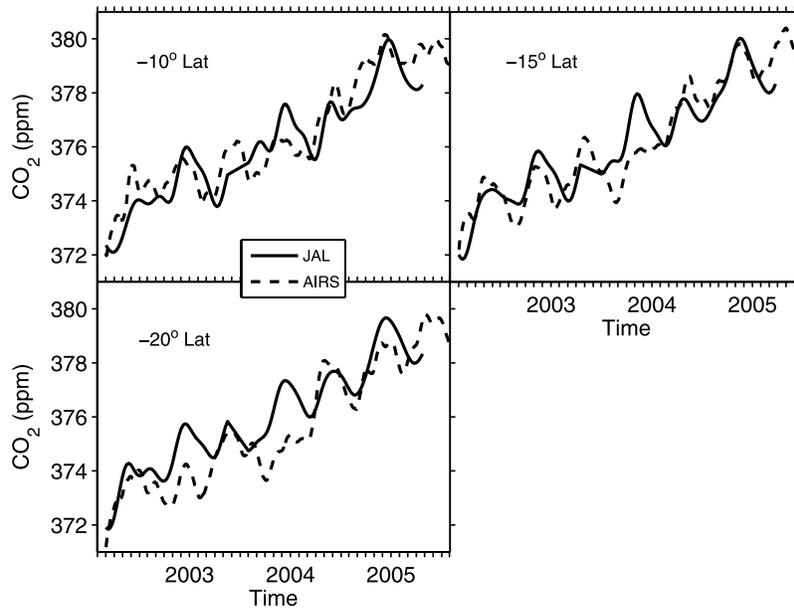


Figure 12. JAL CO₂ measured at 10.5 km (250 mbar) compared with AIRS (LW) for -10° to -20° latitude in 5° increments.

[53] The Rarotonga flights provide a southern hemisphere validation at -21° latitude, shown in Figure 9. Since there is little seasonal variation of CO₂ in the southern hemisphere, this figure highlights the CO₂ growth rate. Note that the y-scale in this figure only spans ~ 8 ppm compared to ~ 15 ppm in Figure 8. The RTA CO₂ is higher than the MBL reference matrix, not unexpected due enhanced mid-tropospheric interhemispherical transport compared to the boundary layer, [see *Strahan et al.*, 1998,

Figure 6]. On average, the RTA values are 0.78 ppm higher than AIRS with a standard deviation of 0.89 ppm.

[54] Figures 10, 11, and 12 compare the 250 mbar JAL CO₂ time series to our AIRS zonal CO₂ from $+30$ to -20° latitude in 5-degree increments. The JAL flights are over open ocean, with little nearby continental influence, especially at these altitudes. The $+30^{\circ}$ to $+15^{\circ}$ latitude graphs show very similar amplitudes and phases for the seasonal cycles of AIRS and JAL. There are differences of up to 2 ppm

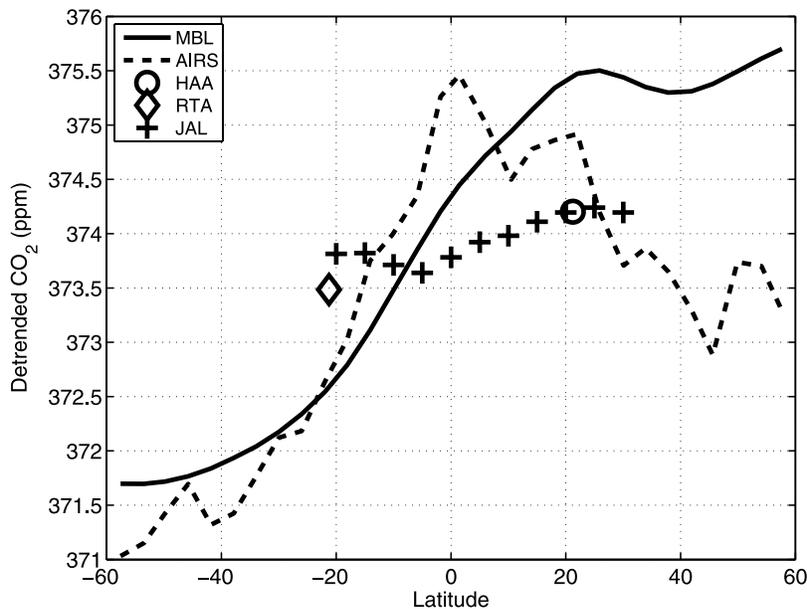


Figure 13. Three-year mean of detrended CO₂ from AIRS (LW) compared with (1) NOAA-ESRL marine boundary layer (MBL) climatology, (2) Molokai Island, Hawaii (HHA) aircraft measurements at 3.5 km (650 mbar), (3) Rarotonga, Cook Islands (RTA) aircraft measurements at 3.5 km (650 mbar), and (4) Japan Airlines (JAL) 10.5 km (250 mbar) measurements. All values are adjusted to a reference time of 1 January 2003.

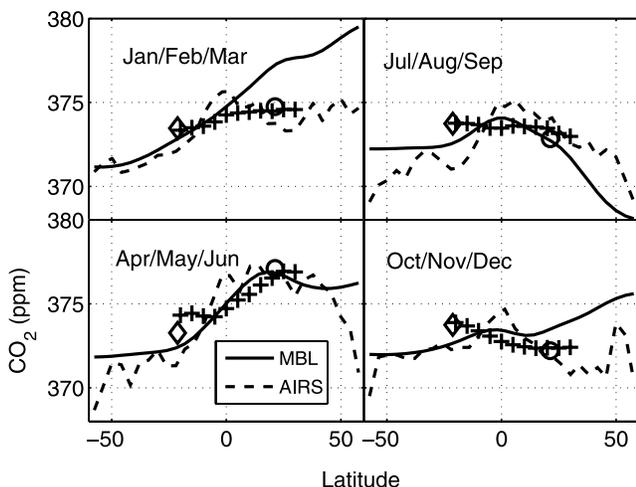


Figure 14. Same data and symbols as in Figure 13 but separated by season.

at these latitudes (early summer 2004/2005 for +30° latitude, for example). In addition, the AIRS CO₂ is slightly higher during 2005. As you move to lower latitudes, the amplitude of the JAL seasonal cycle becomes smaller than the AIRS seasonal cycle. This reduction of the JAL seasonal cycle is most pronounced in Figure 11 for the +10° and +5° latitude data, where the AIRS seasonal cycle starts to lead the JAL cycle. Once you approach the equator, the shapes of the JAL and AIRS seasonal cycles become very similar, with just small phase differences. Below -10° latitude, there are only small oscillations about the overall growth curve of CO₂ for both AIRS and JAL, with no strong correlation other than the yearly growth rate.

[55] Figures 13 and 14 summarize these validation comparisons, and include the MBL climatology for reference. In Figure 13 we detrended and deseasonalized the aircraft time series by fitting them to the following relation,

$$CO_2(t) = C + Rate \times t + \sum_{i=1}^4 [a_i \sin(2\pi i t + \phi_i)] \quad (3)$$

where C is a constant, $Rate$ is the linear growth rate of CO₂, a_i are the amplitudes of the seasonal cycle and the next three harmonics, and ϕ_i are the phases of the seasonal cycle and harmonics. The aircraft points in Figure 13 are $C + Rate \times (t_r - t_o)$ where t_o is the starting time of the aircraft time series, which we set as September 2002 or whenever the aircraft data start after this date. Our reference time is the start of 2003, so $t_r = 2003$. This approach helps remove artifacts due to missing data, although it does suppress any anomalies in these time series, but they are very small.

[56] The AIRS and MBL data are treated slightly differently, since they are basically continuous. For these data we instead construct $CO_2(t) - Rate \times (t - t_r)$ separately for the three years $t = 2003 - 2005$. We then plot the average CO₂ for these three years. This approach leaves any anomalies in the average CO₂, although we find that this only changes the mean (over latitude) CO₂ values by 0.13 ± 0.15 ppm if we instead had plotted the same quantity as for the aircraft data.

[57] Figure 13 clearly shows that the rectifier effect in the Northern Hemisphere produces a mid-troposphere to boundary layer CO₂ gradient above 20° latitude that grows

to ~2 ppm at +60° latitude. In the Southern Hemisphere the AIRS CO₂ generally agrees with MBL to within 1 ppm, with AIRS higher in the tropics and lower in the southern mid-latitudes.

[58] The standard deviations (over time) between the two 3.5 km aircraft flights and the AIRS zonal CO₂ were 0.86, 0.89 ppm respectively for the HAA and RTA sites. Although the JAL flights were at significantly higher altitudes (10 km) the average standard deviation (over time) for the 11 latitudes included in GLOBALVIEW was 1.0 ppm. We did not adjust the AIRS seasonal phase to the in situ CO₂ seasonal phase in computing these standard deviations since it can introduce significant errors at latitudes with small seasonal cycles. For the JAL flights, two latitudes with a seasonal cycle phase almost equal to the AIRS observed phase (20° and 25° latitude) have a standard deviation of 0.9 ppm for all observations, 1.0 ppm for Pacific observations only. Again, these types of comparisons will contain zonal variability, possibly limiting their statistical meaning.

[59] A common way to examine CO₂ variability is to examine the latitudinal dependence by season, as in Figure 14, using the same data processing approach used for Figure 13. In January–March the boundary layer CO₂ is continuing its climb that started in the Fall, but transport to higher altitudes is limited so the AIRS values fall well below the boundary layer values. By April–June the boundary layer has begun to draw down, and the CO₂ profile becomes relatively constant. During the continued draw down in the summer months the Northern Hemisphere boundary layer values drop below the AIRS CO₂ amounts, until the late fall when the boundary layer CO₂ starts to increase again and becomes higher than the AIRS mid- to lower-tropospheric CO₂. In general, the JAL and HAA Northern Hemisphere CO₂ values closely follow the AIRS values over time, although the AIRS CO₂ has a persistent higher value near the equator.

[60] We conclude that the AIRS CO₂ climatology accuracy is approaching the 0.5–1.0 ppm level, at least in the -20° to +30° latitude range where validation data exists. In the southern hemisphere, from -20° to -60° latitude, we have no in situ mid-tropospheric CO₂ data over ocean, and there is always the question if the Northern Hemisphere and tropical ECMWF statistical accuracy we depend upon holds in these lower latitudes.

[61] Finally, we note that close to the equator that the AIRS CO₂ is higher than both the MBL and JAL values. This could be true because of the preferential transport to higher altitudes sensed by the JAL flights, but it could also be due to undetected clouds in the fields of view. This persistent high CO₂ amount does not appear to have a clear seasonal component, suggesting it might not be a cloud effect. Further analysis is needed to completely evaluate these very small deviations that are in the <1 ppm range.

5. Results

[62] Figure 15, top panel, is the main result of this paper, 4 years of zonally averaged CO₂ concentrations at a nominal pressure of 600 mbar. Three features are apparent; (1) the CO₂ concentrations are growing in time, (2) there is a strong seasonal CO₂ cycle in the Northern Hemisphere, and (3) there exists a much weaker seasonal cycle in parts of

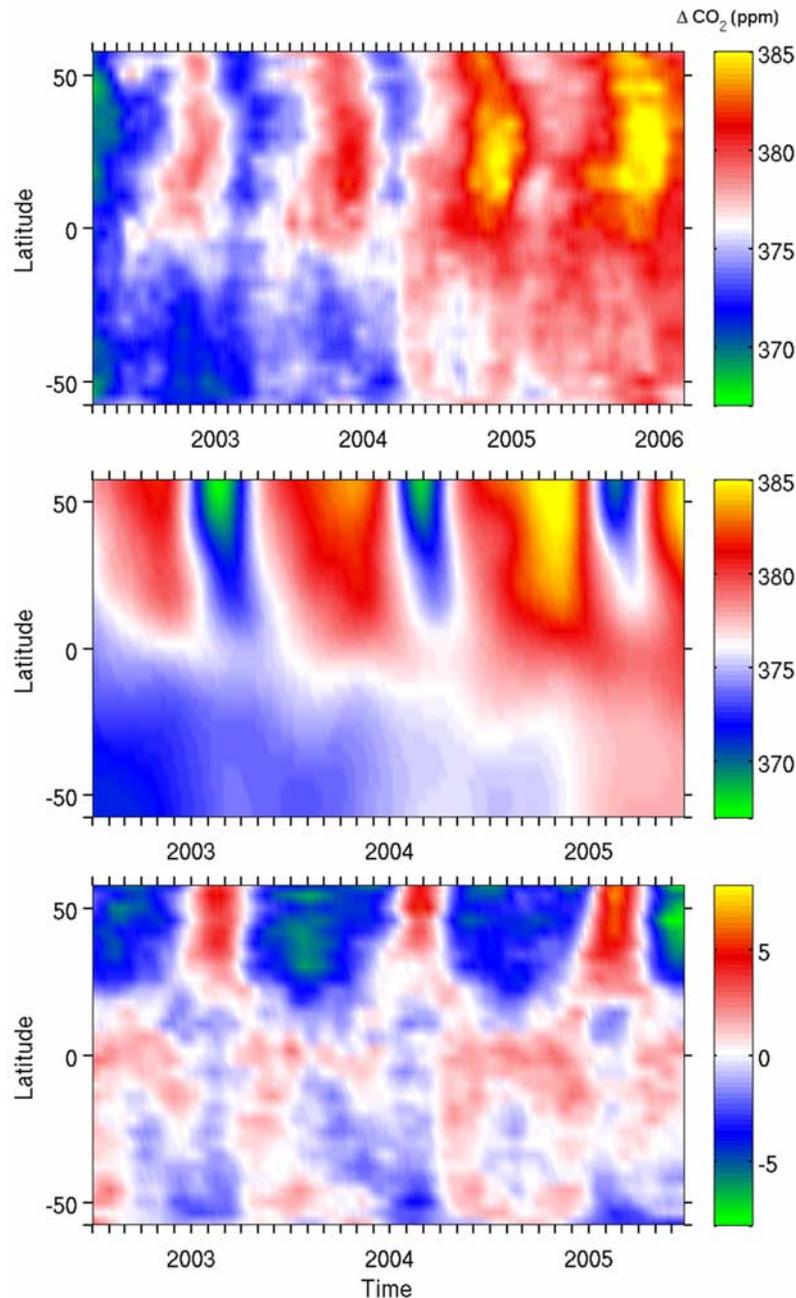


Figure 15. (Top) AIRS retrieved CO₂ using LW channels over the first 4 years of operation. (Middle) NOAA-ESRL MBL CO₂ product for years 2003–2006. (Bottom) AIRS minus MBL CO₂ for years 2003–2006. The AIRS CO₂ nominal peak sensitivity is at 600 mbar, while the MBL CO₂ applies to the marine boundary layer.

the southern hemisphere. CO₂ values vary from a minimum of ~367 ppm to a maximum of ~385, a range of 18 ppm.

[63] Comparisons of our derived zonal CO₂ climatology to the NOAA/ESRL GLOBALVIEW Marine Boundary Layer (MBL) reference matrix provide some level of validation as well as highlighting the differences between our nominal 5 km altitude measurements versus the MBL boundary layer smooth zonal climatology. Figure 15, middle, shows the GLOBALVIEW MBL CO₂ using the same CO₂ scale as in the top, while the AIRS minus MBL difference is shown in the bottom. Note that only 3 years of MBL

CO₂ data is shown since the MBL CO₂ for 2006 was not yet available during this study.

[64] As expected, comparisons of the AIRS to MBL CO₂ shows the strong seasonal rectifier effect [Denning *et al.*, 1996] in the Northern Hemisphere, where the min/max values of the mid-tropospheric seasonal cycle are lower than the boundary layers excursions. The CO₂ rectifier is due to the time covariance between the CO₂ surface fluxes and atmospheric transport. For example, in the late autumn when CO₂ is increasing in the boundary layer, weak, infrequent convection minimizes transport of the seasonally

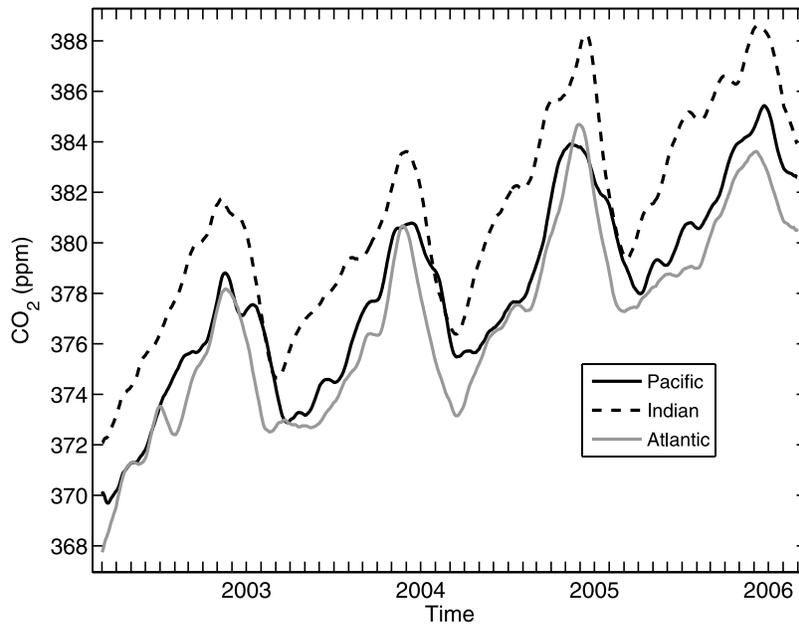


Figure 16. Smoothed CO₂ (LW channels) separated by ocean, averaged over 14°–22° latitude range. Higher CO₂ values in the Indian Ocean are presumably because of transport of nearby Asian continental air.

high CO₂ to the mid-troposphere. Conversely, in the summer months, the boundary layer CO₂ is low due photosynthesis, and increased convection transports these lower CO₂ values to the free troposphere. These so-called rectifier effects lead to accumulation of CO₂ near the ground with depletion aloft in the Northern Hemisphere.

[65] In the southern hemisphere there is evidence for a seasonal cycle nearly 180° out-of-phase with the Northern Hemisphere cycle, but the time series is relatively noisy. The difference between AIRS and MBL appears to have an

anomaly in mid-2005, which will be considered in more detail in section 5.3.

[66] One interesting difference between these two climatologies is seen in the +20 to –10° latitude range in the July time frame. The AIRS CO₂ decreases rather quickly from +20° to –10° latitude, while the MBL CO₂ has a much lower negative slope. The color scale in Figure 15 makes this effect easiest to observe for July 2003. This observation is also evident in our phase analysis of the AIRS and MBL seasonal cycle and is presented in more detail in section 5.1,

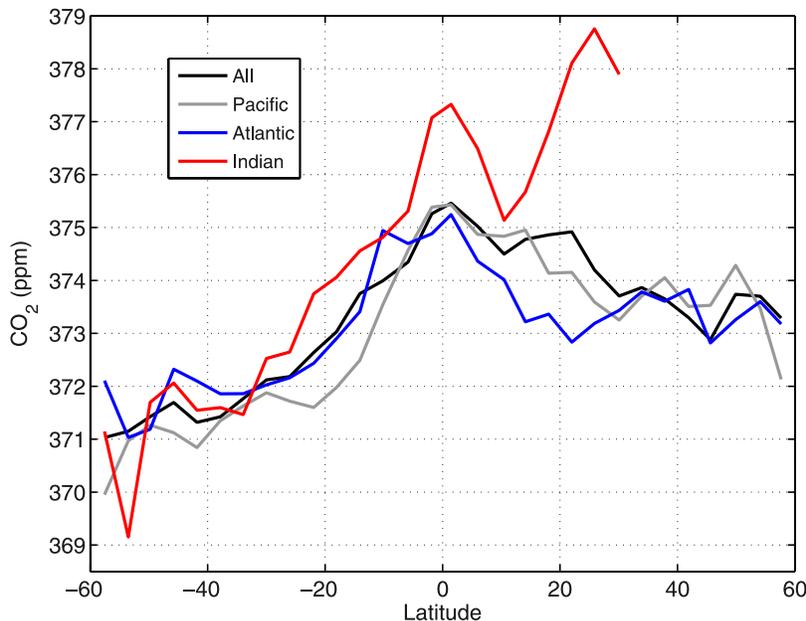


Figure 17. Four-year mean CO₂ from AIRS (LW channels), separated by ocean, plus mean over all oceans. Higher CO₂ values in the northern latitudes of the Indian Ocean are indicative of transport of nearby CO₂-rich Asian continental air.

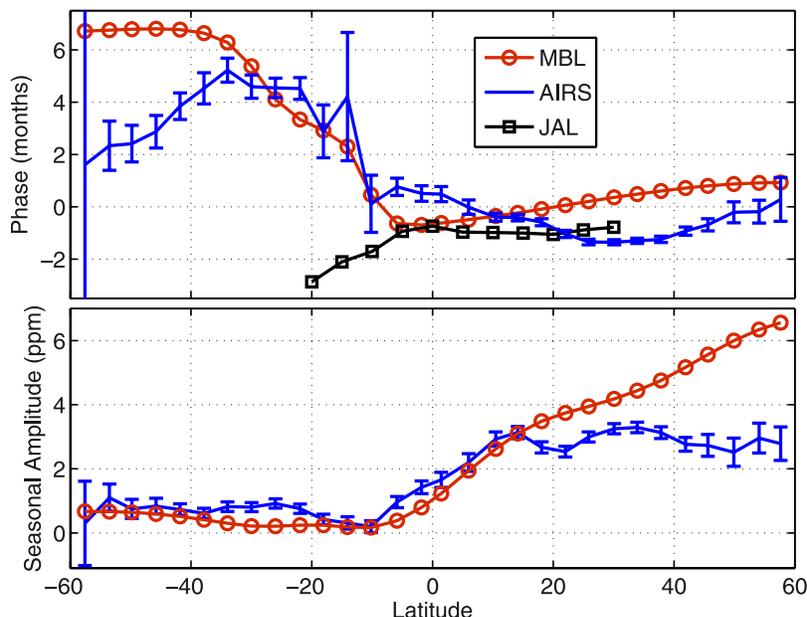


Figure 18. (Top) Phase of the CO₂ seasonal cycle (fundamental mode) for AIRS (LW), NOAA-ESRL MBL, and JAL. Phase is given in months; 2π phase = 12 months. (Bottom) Amplitude of the CO₂ seasonal cycle (fundamental mode) for AIRS (LW) and MBL. Errors bars shown for AIRS in both are 95% confidence intervals.

where we find that the AIRS CO₂ cycle leads the MBL cycle between $+10^\circ$ to -10° latitude. In the higher latitudes the MBL phase leads the AIRS phase because of the time delay in the response of the atmosphere to the surface CO₂ forcing function. We conjecture that this relative phase reversal in the tropics is due to transport of higher latitude, more CO₂ poor air masses, to the tropics during this time frame.

[67] Figure 16 shows our LW 4-year time series of CO₂ averaged from 14° – 22° latitude, separated by ocean. For the Indian ocean this latitude range samples very close to the Asian continent and indeed shows significantly higher CO₂ amounts than the mean CO₂ for the same latitude range for either the Pacific or Atlantic oceans. In Figure 17 we have averaged these CO₂ time series over all four years and plotted the means versus latitude. This plot clearly shows that the Indian Ocean CO₂ values are about 1–4 ppm higher near the continental boundary than in the Atlantic or Pacific. These differences drop sharply as you move to the open Indian ocean at the lower latitudes, where the continental influence has lessened.

[68] We now examine the seasonal cycles of our CO₂ climatology in more detail, mostly relative to NOAA-ESRL’s smoothed boundary layer product, MBL. The seasonal amplitudes and phases presented here are derived from least-squares fits of the raw AIRS CO₂ data, and the MBL time series, to equation (3). The following sections discuss the phase of the fundamental seasonal cycle, ϕ_1 , followed by a discussion of its amplitude, a_1 .

5.1. Seasonal Cycle Phases

[69] The relative phase between the MBL and AIRS CO₂ shown in Figure 18, top, exhibits some interesting behaviors, most likely related to transport. We have plotted the phase in units of months, where 12 months = 2π in phase. Also shown

are the 95% confidence limits from the fits for the AIRS CO₂ seasonal phase.

[70] Phase plots such as these may potentially prove useful for comparisons between transport models and satellite data. This figure should be viewed in parallel with bottom of Figure 18, which shows the amplitude of the seasonal cycle versus latitude.

[71] First we note that the MBL indeed leads the mid-tropospheric AIRS CO₂ in the higher northern latitudes as it takes time for the boundary layer cycle to propagate into the mid-troposphere. However, in the deep tropics the AIRS CO₂ leads MBL between $\pm 10^\circ$. As you move south, the AIRS and MBL phases are similar until -30° latitude where the MBL phase becomes 180° out-of-phase with its northern latitude values, while the AIRS phase starts decreasing as you move south.

[72] The error bars reflect several mechanisms. From $+50^\circ$ to $+60^\circ$ latitude, the error bars are rather large because of the sparsity of data, which introduces both noise, and missing data, into the high-latitude record. In the mid-latitudes the phase error bars are quite small, because the data density increases and the seasonal cycle is quite large. The error bars then increase as you move south mostly because the amplitude of the seasonal cycle is decreasing. Below -10° latitude the retrieved amplitude of seasonal cycle is less than 1 ppm, causing much larger error bars, especially between -10° to -20° latitude. It may also be the case that a harmonic analysis is not the most appropriate way to understand the CO₂ time variability in this latitude region. However, the phase curve is relatively smooth in this region given such small seasonal amplitudes, and it generally follows the MBL phase curve, until -30° latitude where the MBL curve moves steadily higher to reach a level this is 180° out of phase with the northern latitude MBL phase.

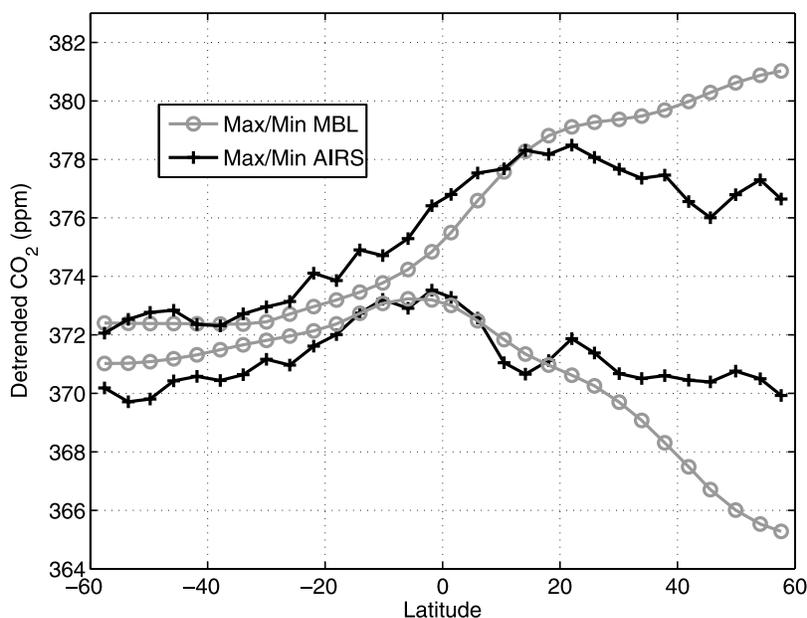


Figure 19. Maxima and minima of the amplitudes of the 3-year detrended AIRS (LW) and NOAA-ESRL MBL time series as a function of latitude. See the text for a possible explanation for the dip in the AIRS minimum CO₂ in the +10° to +20° latitudes.

[73] Below -30° latitude the AIRS CO₂ phase steadily decreases relative to the MBL phase. The error bars increase again as you move south, mostly because of lowered observations statistics. Because the seasonal amplitude is so small in the southern hemisphere, and because there is more uncertainty in the ECMWF model data, the southern hemisphere phase results may be an artifact of our measurement approach, and must be considered with caution. However, it is encouraging that the MBL and AIRS CO₂ phases agree in the -10° to -30° latitudes. Also note that CO₂ may exhibit a 2π phase change slightly below the equator that we cannot detect with these data.

[74] Probably the most interesting phase result is the increase in phase of the AIRS CO₂ relative to MBL between $\pm 10^\circ$. At $+10^\circ$ latitude the AIRS CO₂ starts to lead MBL, reaching a maximum lead of almost $1\frac{1}{2}$ months near -5° . Note that at $+10^\circ$, where the AIRS and MBL phases cross, we also see that the AIRS and MBL seasonal amplitudes become identical, although the mean value (over time) of AIRS is still about 1 ppm lower. This increase in the AIRS CO₂ phase in the deep tropics is likely due to meridional transport from higher latitudes [Nakazawa *et al.*, 1991], especially in the upper tropospheric, which we partially sense with the AIRS CO₂ channels. Note that the AIRS deep tropics CO₂ phase is about the same as the MBL phase at $+30^\circ$.

5.2. Seasonal Cycle Amplitudes

[75] Figure 18, bottom, shows that the AIRS CO₂ seasonal cycle has an almost constant amplitude from $+60^\circ$ to $\sim +10^\circ$ latitude. Also plotted are the 95% confidence limits for the fitted amplitudes. Below 10° , down to our lowest measured latitude of -60° latitude there is very little difference between the AIRS and MBL CO₂ amplitudes. There is some evidence in the -20° to -40° latitude region that the AIRS mid-tropospheric season cycle is larger than

MBL, but the differences are small. This could be due to upper and middle tropospheric interhemispherical transport, which is expected to be larger than in the boundary layer [Nakazawa *et al.*, 1991; Strahan *et al.*, 1998].

[76] The JAL, HAA, and RTA seasonal amplitudes are also plotted in Figure 18, bottom. Both the HAA and RTA amplitudes are very similar to the AIRS CO₂ amplitude, as would be expected, although the RTA amplitude is closer to the MBL amplitude than AIRS.

[77] Since the JAL CO₂ measurements are taken at 250 mbar, well above the ~ 550 mbar peak of the AIRS CO₂ sensitivity, we do not expect the amplitudes to be identical. In general one would expect the JAL amplitude to be smaller than both MBL and AIRS in the northern latitudes since it is more distant from the CO₂ forcing function. However, from -10° to -15° latitude the JAL amplitude is considerably higher than either AIRS or MBL, neither of which show significant seasonal variability. This is also consistent with enhanced meridional transport from the Northern Hemisphere at higher altitudes [see Strahan *et al.*, 1998, Figure 6].

[78] Between -20° and -40° latitude the amplitude of the AIRS seasonal cycle is almost 1 ppm, compared to the MBL seasonal cycle amplitude of close to zero. This result would again suggest that meridional transport of air from the Northern Hemisphere may be dominating the CO₂ cycle in the nominal 300–800 mbar region sensed by our AIRS measurement. No JAL data exists for comparison between -20° and -40° latitudes.

[79] Figure 19 is another view of the AIRS and MBL seasonal amplitude, except here we plot the maximum and minimum of the seasonal cycle over the whole year. This view of the data highlights differences between MBL and AIRS during the draw-down of CO₂ versus the build-up of CO₂. Note that the rectifier effect is very symmetric at the highest latitudes. As you move south, it appears that during

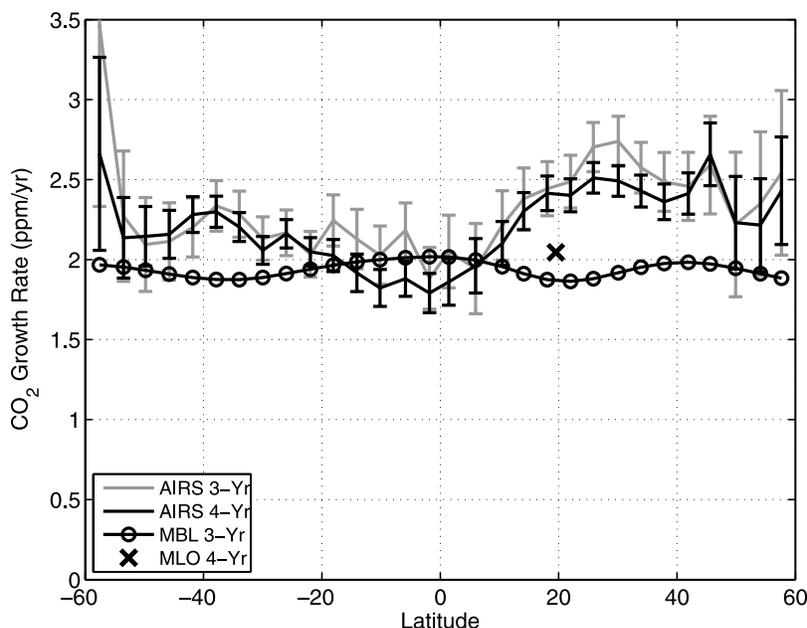


Figure 20. CO₂ growth rates for the full 4-year AIRS record, for the 2003–2005 3-year AIRS record (both using LW channels), and for the same 2003–2005 3-year record from the NOAA-ESRL MBL climatology. Also shown in the Mauna Loa is the growth rate for the same time period as the AIRS 4-year CO₂ record. Error bars shown for the AIRS measurements are 95% confidence intervals.

CO₂ draw-down (minimum) the AIRS mid-tropospheric CO₂ reaches or goes slightly below the MBL minimum from +10° to +20°. This may be caused by the fact that the sharp Northern Hemisphere draw-down of CO₂ begins in May when convection is increasing. CO₂ transport from higher latitudes pushes the +10° latitude mid-tropospheric CO₂ downward more quickly than the boundary layer values at +10°, which are also decreasing, but not as quickly. This negative dip in the AIRS CO₂ minimum is what causes the slight increase in the CO₂ seasonal amplitude at this latitude. A corresponding increase in the maximum of the AIRS CO₂ near +10° latitude is not expected since there is less convection in the winter and early spring when the Northern Hemisphere CO₂ is reaching a maximum.

[80] The MBL seasonal oscillation in the southern hemisphere is very small, generally less than 1 ppm. The AIRS-derived maximum CO₂ in this region is very close to the MBL values, while the AIRS minimum values are about 2 ppm lower than the AIRS maximum values. As discussed earlier, these very small differences between AIRS and MBL in the southern hemisphere are difficult to validate, and may be due to errors in either data set. However, they both show similar trends, both with latitude and with the offset between maximum and minimum values.

5.3. CO₂ Growth Rates

[81] The CO₂ growth rates as a function of latitude, fitted using equation (3), are plotting in Figure 20, for (1) the AIRS 4-year CO₂ record, (2) three years of AIRS, 2003–2005, (3) MBL during the same 3-year time span, and (4) the 4-year Mauna Loa rate. Deriving accurate CO₂ growth rate information from AIRS is highly dependent on the stability of both the AIRS radiometry and the ECMWF model. These rates are averages from 1 September 2002 to

30 August 2006 for the 4-year rate, and from 1 January 2003 to 31 December 2005 for the 3-year rates. The AIRS 4-year rates include the February 2006 time period when we observed a change in the ECMWF model that was reflected as an offset of ~3 ppm in the SW versus LW AIRS CO₂ amounts. The 3-year rates do not include the time period of this model change.

[82] The AIRS CO₂ growth rates were derived using the raw daily average CO₂. For the –50° to +50° latitudes, almost all daily bins are populated, allowing a good estimation of the level of autocorrelation in each of the zonally binned time series. We used the lag-1 autocorrelation to determine if the normal least-squares error estimates must be increased to account for non-Gaussian statistics [Santer *et al.*, 2000] and found these corrections to be negligible for the ±40° latitude range where our daily time series is continuous. Consequently, the rate error bars are the standard least-squares 95% confidence intervals.

[83] In general the retrieved rates are quite close to the equivalent MBL rates, with a maximum difference of about 0.5 ppm/year in the 20°–40° latitude range. Note that 0.5 ppm/year is equivalent to only a 0.015 K/year brightness temperature change in the 791.7 cm⁻¹ channel. The AIRS CO₂ rates are about 0.5 ppm/year higher for +20° latitude and beyond, and about 0.25 ppm/year higher in the southern mid-latitudes, from ~–20° to –55° latitude. Our error bars grow rapidly as you approach ±60° latitude because of far fewer clear FOVs in our data set, with many days missing, especially in the winter. In the tropics from ~+10 to –20° latitude the AIRS and MBL rates agree within the AIRS error bars.

[84] Table 1 summarizes the rates measurements and comparison to other sources. If one averages over all latitudes, the AIRS 4-year rate is 2.21 ppm/year with a standard deviation of ±0.24 ppm/year (standard deviation is over

Table 1. Growth Rates From Various Data Sets^a

Time Series	Rate/Std, ppm/yr
AIRS 4 yr	2.21 ± 0.24
AIRS 3 yr	2.32 ± 0.31
MBL 3 yr	1.94 ± 0.05
MLO 4 yr	2.05
AIRS 20–40° subset	(1.86, 2.56) = 2.21
MBL 20–40° subset	(2.07, 2.88) = 2.48

^aThe AIRS growth rates all use the LW CO₂. Standard deviations are taken over latitudes. Rates averaged from only 20° to 40° latitude are only for two particular time period subsets that are noted in the text and in the Figure 21 caption.

latitude). The AIRS 3-year rate is 2.32 ± 0.31 ppm/year. This compares to the mean MBL rate of 1.94 ppm/year (3-year rate). The difference between the AIRS and MBL 3-year rates is 0.38 ppm/year, which is equivalent to a brightness temperature change of 0.011 K/year. The MLO 4-year rate differs from the latitudinally averaged AIRS 4-year rate by 0.16 ppm/year, or a brightness temperature difference of 0.0048 K/year. If you just compare the AIRS 4-year rate at the MLO latitude, to the MLO 4-year rate, you get a difference of 0.36 ppm/year or 0.011 K/year in brightness temperature. These results suggest that the AIRS CO₂ growth rates are accurate to at least 0.4 ppm/year, and possibly better.

[85] Some insight into the differences between the AIRS versus MBL rates is gained by plotting the deseasonalized time-series. Figure 21 is a graph of

$$CO_2(t) - C - \sum_{i=1}^4 [a_i \sin(2\pi it + \phi_i)] \quad (4)$$

which subtracts the fitted offset constant and sinusoidal terms from the raw CO₂ time series. We chose to plot the

mean of this series between 20° and 40° latitude for the LW and SW AIRS CO₂, and for MBL, where we have the largest disagreement between AIRS and MBL. This plot clearly indicates that the AIRS CO₂ diverges from MBL in late 2004, with agreement appearing to return at the end of 2005. We now concentrate on the time period between the two vertical bars in this figure. It appears that in late 2004, the MBL growth rate almost stopped for several months, while at the same time the AIRS growth rate increased above normal levels. This produced an offset between MBL and AIRS during most of 2005 of almost 2 ppm.

[86] The reasonably good agreement between the SW and LW CO₂ growth rate shown in Figure 21 (before the February 2006 ECMWF change) suggests that the difference between AIRS and MBL is not due to our use of ECMWF for the atmospheric temperature. (This figure does show the relatively large disagreement between the SW and LW AIRS CO₂ starting in February 2006.) A linear least-squares fit to these curves for the growth rates between 1 January 2003 and the left-hand side vertical bar, and a separate fit from the right-hand side vertical bar to 31 December 2005 are shown in Table 1 under AIRS/MBL 20–40° latitude subset. The second column shows the fitted rates for each time series before and after the vertical bars, along with their mean values. We observe that both of the AIRS rates shown are *lower* than the MBL rates, with an average AIRS CO₂ growth rate of 2.21 ppm/year compared to the MBL rate of 2.48 ppm/year. Thus we conclude that the higher rates for AIRS than MBL shown in Figure 20 are mostly due to very different behavior in these two time series during the late fall of 2004. This result increases our confidence in the radiometric stability of AIRS.

[87] The differences between the MBL and AIRS CO₂ growth rates are very small, and diagnosing these observed

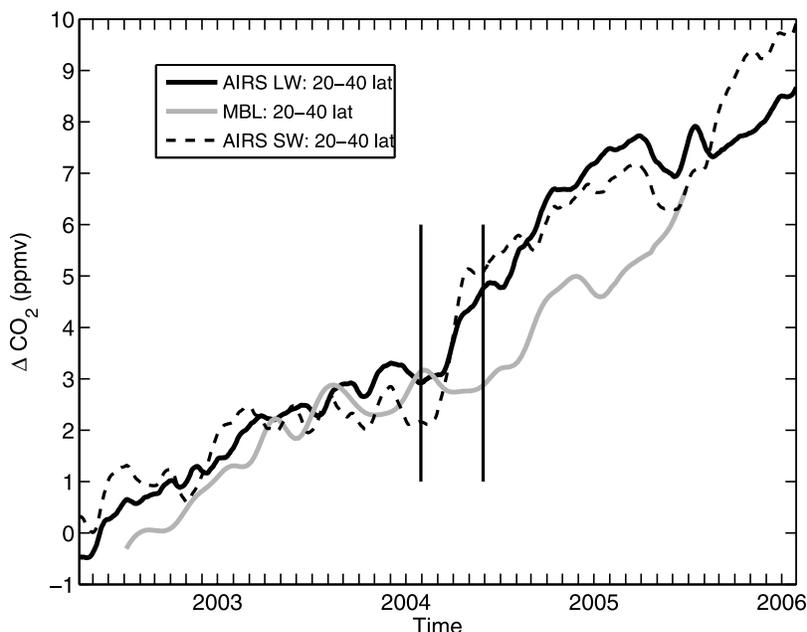


Figure 21. The AIRS LW and SW CO₂ record and the NOAA-ESRL MBL record using deseasonalized data, averaged over 20° to 40° latitude. The two vertical bars indicate a time span when the growth of CO₂ in the oceanic boundary layer (MBL) was significantly different than that in the AIRS-measured CO₂ growth rates for both LW and SW. Table 1 gives the growth rates for AIRS and MBL for times previous to the first vertical bar, times after the second vertical bar, and their average.

small differences is beyond the scope of this paper. It must be kept in mind that these are zonally averaged rates, and both the AIRS and MBL rates are highly likely to weight points differently in the longitude mean. For example, Figure 1 shows that the longitude of clear FOVs varies greatly with latitude. The mean longitude of our observations (using -180° to $+180^{\circ}$) varies within $\sim\pm 30^{\circ}$ latitude for all latitudes except for the two zones within $\pm 2^{\circ}$ latitude of the equator, which have a mean longitude of $\sim -50^{\circ}$. Our zonal ocean CO₂ may also have a significant continental influence in the Indian/Arabian sea, where we found a large number of clear FOVs.

[88] A possible source of error for the AIRS CO₂ rates is drift in the AIRS radiometric calibration. Aumann *et al.* [2006] have attempted to address this in the context of validation of the AIRS radiometric accuracy, where he estimated that AIRS is stable to at least 0.016 K/year with a 95% confidence limit, now estimated to be 0.010 K/year by H.H. Aumann (unpublished data, 2007). He used the NOAA RTG-SST sea surface product [Thiébaux *et al.*, 2003] in the tropics as a known temperature to compare to AIRS retrieved SST, using essentially the same clear-FOV subset that we used in this work. His AIRS SST uses the 2616 cm⁻¹ channel, which is the most transparent AIRS channel and has essentially no absorption because of CO₂.

[89] We repeated his analysis using our data subset, and the ECMWF SST product, which is stated by ECMWF to be the NCEP SST analysis. Comparisons for one day in 2003 confirm that the NCEP and ECMWF SST are essentially identical. Our subset gives a drift relative to the ECMWF tropical SST of 12 mK \pm 15 mK (taking into account autocorrelation of time series), essentially the same result. However, as noted by Aumann *et al.* [2006], the RTA SST record contains an offset in May 2004. We find that our Obs-Calcs with respect to ECMWF SST is +4 mK/year for Sept. 2002 through April 2004, and -6 mK/year for June 2004 through August 2006, both of which are smaller than the drift from the combined 4-year data set which includes the May 2004 RTG SST offset.

[90] This analysis suggests that the AIRS radiometry is stable to at least 10 mK/year, implying a maximum uncertainty of 0.3 ppm/year in the AIRS CO₂ growth rate. At this time, we believe that it is not possible to associate differences in the AIRS and MBL growth rates to any known drift in the AIRS radiometry. In addition, it is highly unlikely that a radiometric drift could cause the latitudinal variability of the CO₂ growth rates shown in Figure 20.

6. Summary and Conclusions

[91] A 4-year zonal CO₂ climatology derived from AIRS for ocean-only measurements was shown to be accurate to ~ 0.5 –1 ppm, at least in the -20° to $+30^{\circ}$ latitude region where validation data exists. Comparisons to the NOAA GLOBALVIEW MBL CO₂ smoothed product clearly shows the operation of rectifier effect, which limits venting of CO₂ out of the boundary layer in northern latitudes. Comparisons of the phase between the MBL and AIRS CO₂ seasonal cycles may offer some insights into the transport of CO₂ from the northern to southern latitudes. In particular we find that in the deep tropics the AIRS CO₂ leads the MBL CO₂ by $\sim 1\frac{1}{2}$ months, presumably because of transport from

higher latitudes where the CO₂ seasonal cycle leads the tropical MBL phase.

[92] The observed CO₂ growth rates during the three years 2003–2005, averaged over all latitudes, agree with the MBL growth rates to within 0.38 ppm/year. However, our CO₂ rates are systematically higher than the MBL rates in the mid-latitudes by 0.3–0.5 ppm/year. This appears to be due to some event, or unknown error source, that was in force during late 2004 when the MBL and AIRS CO₂ rates diverged for a short time period.

[93] This work establishes the ability of AIRS to measure global CO₂ concentrations in the mid- to lower-troposphere with high accuracy. By using only very clear FOVs over ocean, our CO₂ climatology is largely unaffected by clouds, which allowed us to use such low-peaking CO₂ channels. Unfortunately, there are limited lower to middle tropospheric validation measurements over ocean; we used two sets of 3.5-km aircraft measurements to partially validate our CO₂ results, with additional partial validation from the more extensive 250 mbar measurement aboard the Pacific JAL airline flights.

[94] There are also questions of how sampling may bias our results. The use of clear only FOVs generally means we have avoided regions of strong convection that might prove interesting for transport studies. In addition, some of our zonal averages come from regions without strong nearby continental flow, while others region might be strongly influenced by continental outflow.

[95] Future studies will include detailed model comparisons, retrievals over land, and possibly the use of AIRS cloud-cleared radiances for the CO₂ measurement. This last possibility could potentially allow more detailed validation by greatly increasing the density of usable data, and hopefully provide enough data to generate meaningful maps, which will help alleviate a sampling bias and possibly provide better transport information as well as information on source and sink regions. Increased data density should also allow us to generate zonal averages from longitudinally gridded data, which would be more statistically representative. In addition, given the potential accuracy of the results presented here, more detailed studies of the effects of residual cloudiness on our derived CO₂ are warranted. This work does, however, appear to represent a vast amount of new global CO₂ data for the mid- to lower-troposphere that will hopefully prove useful to the carbon modeling community.

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