Carbon Dioxide Laser Absorption Spectrometer (CO2LAS) aircraft measurements of CO_2

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ABSTRACT

The Jet Propulsion Laboratory Carbon Dioxide Laser Absorption Spectrometer (CO2LAS) utilizes Integrated Path Differential Absorption (IPDA) at 2.05 µm to obtain CO_2 column mixing ratios weighted heavily in the boundary layer. CO2LAS employs a coherent detection receiver and continuous-wave Th:Ho:YLF laser transmitters with output powers around 100 milliwatts. An offset frequency-locking scheme coupled to an absolute frequency reference enables the frequencies of the online and offline lasers to be held to within 200 kHz of desired values. We describe results from 2009 field campaigns when CO2LAS flew on the Twin Otter. We also describe spectroscopic studies aimed at uncovering potential biases in lidar CO_2 retrievals at 2.05 µm.

Keywords: Lidar, heterodyne, laser

1. INTRODUCTION

Differential absorption, heterodyne detection remote sensing of atmospheric trace gases using lasers has been described since the early 1970s.[1] The heterodyne technique generates signals far removed from most noise sources which enables detection of backscattered radiation at return powers less than 10^{-15} W for receivers designed near 2 µm. This permits low-power (sub-Watt) lasers to act as transmitters and thus there is great potential for low-cost, long-endurance, low Earth orbit remote sensing of greenhouse gases with this technique.

The CO_2 Laser Absorption Spectrometer (CO2LAS)[2] is a differential absorption, heterodyne detection remote sensing instrument developed jointly by JPL and Lockheed Martin Coherent Technologies. It is intended to provide the next generation of active CO_2 monitoring from space as part of NASA's ASCENDS mission.[3] CO2LAS will do this by measuring averaged, dry-mole-fraction column CO_2 (X_{CO2}) weighted heavily in the lower troposphere.

Precision requirements for satellite-borne X_{CO2} retrievals are extremely demanding - on the level of a ppm or better.[4] It is essential that sources of bias be understood and eliminated wherever possible. Temporal and geographic biases of a ppm can effect PgC changes in how we proportion Earth's carbon sources and sinks.[5]

We present results from field experiments conducted in 2009 and discuss instrument precision and a positive offset in CO_2 retrievals. We analyze the contribution of incomplete spectroscopic modeling to the offset.

2. INSTRUMENT

Instrument specifications are given in Table 1 and a schematic is shown in Figure 1. CO2LAS uses three diode-laser pumped continuous-wave Tm,Ho:YLF lasers denoted reference, online, and offline. The reference laser is locked to the center of the $^{12}CO_2$ $20^01_3 \leftarrow 00^00$ R30 line (4875.749 cm^{-1}). The online laser (4875.882 cm^{-1}) is in turn locked to the reference laser using a high bandwidth receiver. The offline laser (4875.225 cm^{-1}) is locked to the reference laser after first shifting its frequency with an EOM. Online and offline transmitted powers are monitored. Portions of online and offline beams are split off to act as the local oscillator for the return signals.

Table 1. CO2LAS specifications.

Parameter	**Value**
CO_2 line-locked frequency	4875.749 cm^{-1}
Online frequency	4875.882 cm^{-1}
Offline frequency	4875.225 cm^{-1}
Laser output power	100 mW
Transmit/Receive apertures	10 cm diameter
FOV	60 µrad
Photomixer type	InGaAs
Digitization	14 bits/50 MHz

The laser beams are telescoped and then transmitted through a downward-facing window. Online and offline light are differentially attenuated by atmospheric CO_2. Figure 2 shows their frequencies with respect to the atmospheric transmission profile. As the beams scatter off the Earth's surface, some scattered light is captured by the same telescopes from which they were transmitted. Since pointing is off-nadir, the frequency of the return light is Doppler shifted tens of MHz. The return beams beat against the local oscillator to produce online and offline heterodyne signals.

Time-domain heterodyne signals are then fast-Fourier-transformed (FFT) into frequency-domain signals. An online channel FFT signal is shown in Figure 3. A similar signal for the offline channel is also acquired. Signals are normalized with respect to transmitted power and are then further processed accounting for aircraft pitch, Shuttle Radar Topography Mission topography (30 m resolution), and humidity, pressure, and temperature profiles.

Ultimately, the quantity retrieved by CO2LAS is the differential absorption optical depth (DAOD)

$$\mathrm{DAOD} = \ln(\tau_{off}/\tau_{on}) = 0.5\times\ln(P_{off}/P_{on}) \tag{1}$$

where τ_{on} and τ_{off} are atmospheric transmission at the online and offline frequencies and P_{on} and P_{off} are received power at the online and offline frequencies. DAOD is then translated into X_{CO2} via forward modeling using the Line-By-Line Radiative Transfer Model (LBLRTM).

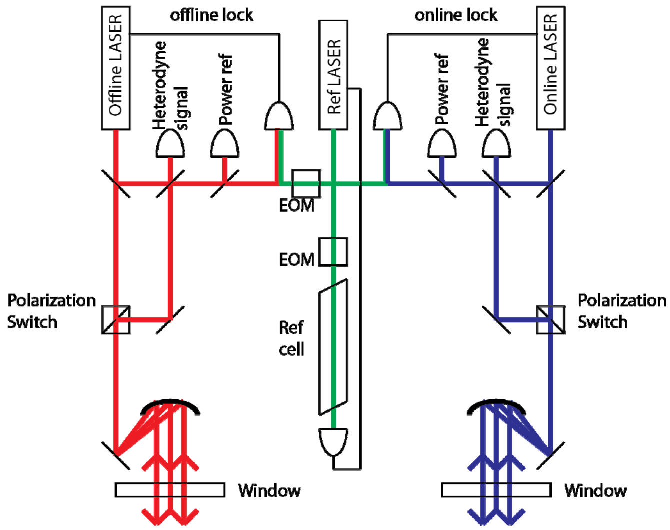

Figure 1. Instrument schematic. EOM = electro-optic modulator.

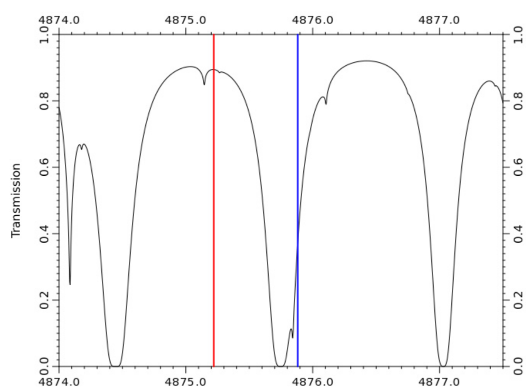

Figure 2. Modeled atmospheric transmission from low Earth orbit. Red and blue lines are offline and online frequencies, respectively.

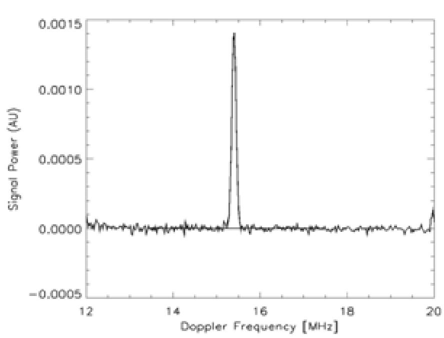

Figure 3. Periodogram with noise floor removed.

3. FIELD MEASUREMENTS

Field experiments were conducted at El Mirage dry lakebed, CA, April 2009 (2 flights, 3 altitudes each flight), and over the DOE ARM-SGP site, OK, July-August 2009 (4 flights, 4 altitudes each flight). For these experiments, retrieved CO_2 was greater than true column CO_2 by a factor of 1.4. Since the offset was constant between the El Mirage and Oklahoma experiments and since there was no indication that it was affected by surface or atmospheric conditions, we subtracted it out - these adjusted measurements constitute our reported X_{CO2} retrievals.

3.1 Signal fluctuations

Figure 4 shows X_{CO2} retrieved by CO2LAS during an overpass at 3 km elevation near the ARM-SGP site in Oklahoma August 31, 2009, 11:30 to 11:40 local time. Fluctuations in X_{CO2} are ±8 ppm peak-to-peak around a mean value of 382 ppm for integration time of 10 s. The fluctuations are instrumental in nature - true X_{CO2} variability was less than 2 ppm inferred from in situ measurements made nearby.

Most of the variability seen in Figure 4 is due to speckle fluctuations. For this integration time, speckle noise is ±5 ppm. Noise due to speckle over the entire 35 km overpass (akin to the standard error) is 0.6 ppm.

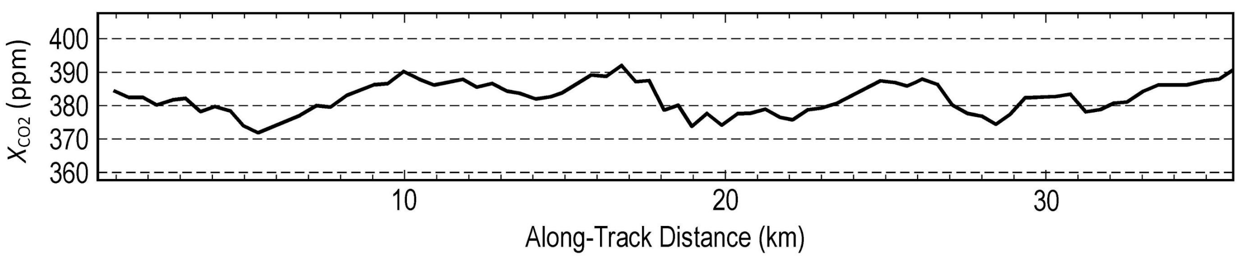

Figure 4. CO2LAS X_{CO2} during August 31, 2009 Oklahoma experiment, 3 km elevation. 11:30 to 11:40 local time.

3.2 Offset

Currently, we have identified two potential sources of the offset. The one considered most significant is the result of a range dependent heterodyne efficiency[6] which may originate from slight differences in online and offline receiving efficiencies. An important characteristic of this behavior is that it decreases with altitude. If this is indeed responsible for the offset observed during our aircraft flights, it is likely such an offset will not be present in low Earth orbit retrievals.

The other potential cause of offset may be due to inaccurate spectroscopic modeling. This is discussed below.

4. SPECTROSCOPY

Often, spectroscopic modeling of Earth's atmosphere describes individual optical transitions using the Voigt lineshape. The Voigt lineshape convolves a Lorentzian description of pressure broadening with Doppler broadening. It is conceptually simple, has been exhaustively studied, and many numerical algorithms have been developed for it. These algorithms are relatively fast and have been tested for decades.

It is recognized that the Lorentzian description is only an approximation. Better descriptions of lineshape assimilate more detailed views of inter- and intra- molecular processes like collisions and quantum-state interactions. One such description incorporates linemixing[7] which has been shown to be important for atmospheric CO_2 retrievals at 2 μm.[8]

For instruments retrieving X_{CO2} from space, the Voigt lineshape is likely inadequate due to the demanding precision and accuracy requirements. In the case of instruments, like CO2LAS, that do not fit entire lineshapes or band profiles but rather contrast light attenuation at selected sounding frequencies, use of an inappropriate lineshape can cause even greater errors. This is exemplified in Figure 5 where we compare a spectroscopic model of light transmitted through Earth's atmosphere with and without linemixing. At the 1 percent level, it is difficult to distinguish between models in the spectral region where most 2.05 μm lidar instruments propose to retrieve their offline frequency values (Figure 4, middle). However, at the spectral region where CO2LAS has its online frequency, the difference between models can be easily seen at the 1 percent level (Figure 4, bottom). It is important to note that this frequency was chosen because it is the best balance between high sensitivity to tropospheric CO_2 and good signal-to-noise for low Earth orbit retrievals.[9] It is also important to note that the issue described here applies to the opposite side of the $^{12}CO_2$ $20^01_3 \leftarrow 00^00$ R30 line which is another frequency region sometimes cited for online measurements.[10]

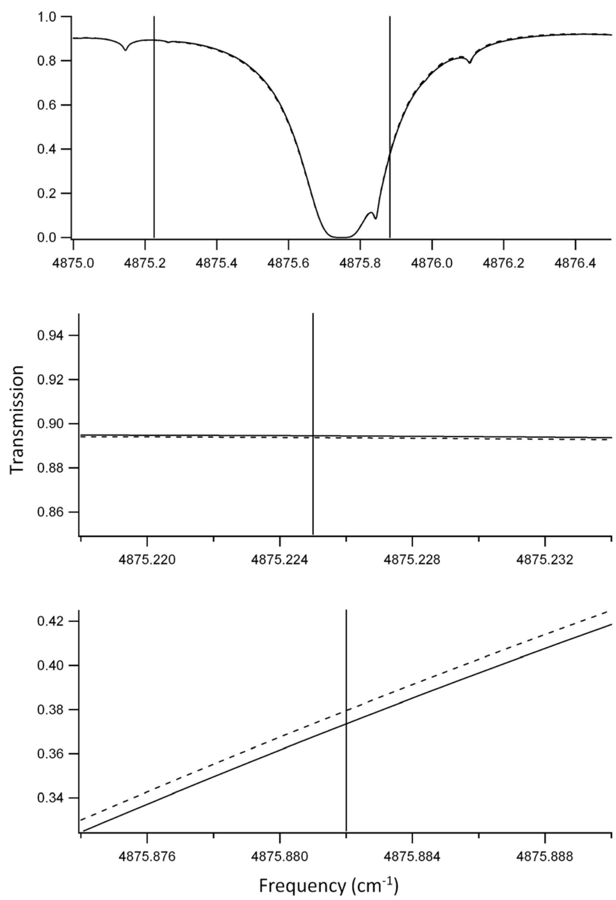

Figure 5. Modeled transmission from low Earth orbit contrasting linemixing and non-linemixing models. The vertical lines are CO2LAS online and offline frequencies. Top: Whole region around the $^{12}CO_2$ $20^01_3 \leftarrow 00^00$ R30. Middle: Close-up of offline frequency. Bottom: Close-up of online frequency.

We first sought to confirm the effect of linemixing on CO_2 spectroscopic measurements near 2.05 μm. Experiments were conducted between 50 and 1000 mbar at room temperature. A 5.03% mixture of CO_2 in dry air was flowed through an Aerodyne 36-m optical pathlength multipass cell. A room-temperature, continuous-wave diode laser emitting at 2.05 µm (Nanoplus GmbH, < 30 dB SMSR, > 2 mW) was used to probe absorption by the gas mixture as a function of frequency.

Data acquired at 840 mbar is shown in Figure 6. This concentration of CO_2 is low enough to let us assume most broadening effects were due to air-CO_2 interaction but high enough to allow us to obtain a spectrum that reasonably represents what a low Earth orbit instrument might observe, with transmissions approaching zero at the peaks.

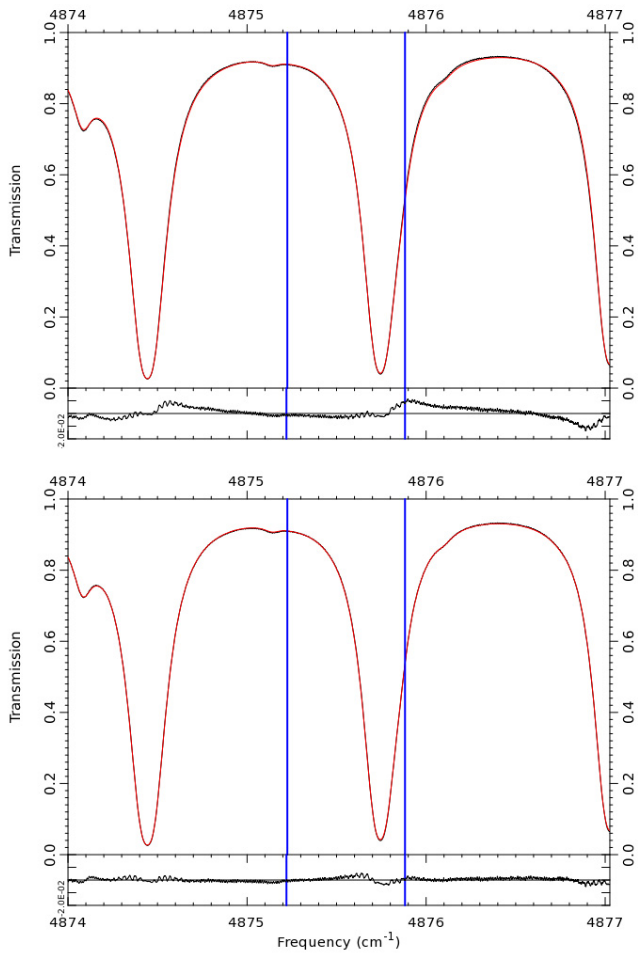

Figure 6. Fits to laboratory data at 840 mbar. Non-linemixing (top) and linemixing (bottom).

We then fit the data in Figure 6 with a spectroscopic model using linemixing and non-linemixing (red lines). It is clear from the residuals that the linemixing model is significantly more accurate that the non-linemixing model. We conducted these experiments at nine different pressures, over eleven $^{12}CO_2$ $20^01_3 \leftarrow 00^00$ transitions (16 cm^{-1}) and measured linemixing coefficients (Y_ℓ) for each of these eleven lines (ℓ denotes an individual line index). Our measurements agree with linemixing coefficients for the $^{12}CO_2$ $30^01_3 \leftarrow 00^00$ and $30^01_2 \leftarrow 00^00$ bands.[8, 11-13]

We then used the linemixing model to contrast forward-modeling using linemixing against not-using linemixing assuming the U.S. Standard atmosphere, 0% humidity, and $[CO_2]$ homogeneously mixed. For a CO2LAS instrument at 40 km meters above sea level (masl), ranging all the way to sea level, assume the measured DAOD is 0.8551. The linemixing model would interpret this is X_{CO2} = 390.0 ppm while the non-linemixing model would interpret this as 382.4 ppm. There is a difference of 7.6 ppm between the models.

This discrepancy is not merely an offset. The effects of linemixing are non-linear with respect to pressure, becoming more important at higher pressure. The CO2LAS weighting function is skewed toward those conditions for which linemixing is most important. We tested this elevation bias by contrasting models in which we ranged to sea-level versus models in which we ranged to the Tibetan Plateau. These tests are difficult to interpret because one model needs to be calibrated against the other (to remove the offset discussed in the preceding paragraph). In the present case, we assumed true X_{CO2} would be acquired by aircraft/ground-based measurements at Park Falls, WI, elevation 560 masl. We then scaled the $^{12}CO_2$ $20^01_3 \leftarrow 00^00$ R30 linestrength in the non-linemixing model such that DAOD between non-linemixing and linemixing model match at Park Falls. Then we compared DAODs at 0 masl and 4500 masl (e.g. Tibetan Plateau). We found a +0.7 ppm bias going from 0 masl to 4500 masl assuming this method of calibration.

5. CONCLUSIONS

At JPL, differential absorption, heterodyne detection remote sensing started as a concept,[1] evolved to a first-generation aircraft instrument,[14] and now is a second-generation aircraft instrument[2]. We examine recent flight and laboratory experiments that increase the ability of active remote sensing to quantify Earth's carbon sources and sinks.

Acknowledgements

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