Field Evaluation of Column CO₂ Retrievals From Intensity-Modulated Continuous-Wave Differential Absorption Lidar Measurements During the ACT-America Campaign

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Abstract We present an evaluation of airborne intensity-modulated continuous-wave (IM-CW) lidar measurements of atmospheric column CO₂ mole fractions during the Atmospheric Carbon and Transport–America (ACT-America) project. This lidar system transmits online and offline wavelengths simultaneously on the 1.57111-µm CO₂ absorption line, with each modulated wavelength using orthogonal swept frequency waveforms. After the spectral characteristics of this system were calibrated through short-path measurements, we used the HITRAN spectroscopic database to calculate the average-column CO₂ mole fraction (XCO₂) from the lidar-measured optical depths. Using in situ measurements of meteorological parameters and CO₂ concentrations for calibration data, we demonstrate that our lidar CO₂ measurements were consistent from season to season and had an absolute calibration error (standard deviation) of 0.80 ppm when compared to XCO₂ values calculated from in situ measurements. By using a 10-s or longer moving average, a precision of 1 ppm or better was obtained. The estimated CO₂ measurement precision for 0.1-, 1-, 10-, and 60-s averages was determined to be 3.4, 1.2, 0.43, and 0.26 ppm, respectively. These correspond to measurement signal-to-noise ratios of 120, 330, 950, and 1,600, respectively. The drift in XCO₂ over 1-hr of flight time was found to be below 0.1 ppm. These analyses demonstrate that the measurement stability, precision, and accuracy are all well below the thresholds needed to study synoptic-scale variations in atmospheric XCO₂.

1. Introduction

Atmospheric carbon dioxide (CO₂) is a crucial part of the Earth’s carbon cycle and one of the major greenhouse gases (GHGs) in the Earth’s climate system. The CO₂ concentration within the atmosphere has significantly changed over the last 150 years, due mainly to anthropogenic activities. Understanding the carbon cycle is essential for diagnosing current, and predicting future, climate change (Gregory et al., 2009; Marquis & Tans, 2008; Michalak et al., 2011). The Earth’s terrestrial biosphere has been a strong net sink of atmospheric CO₂ for decades (e.g., Le Quéré et al., 2009), substantially slowing the rate of accumulation of CO₂ in the atmosphere from combustion of fossil fuels. The causes of the net biogenic CO₂ sink, its location and magnitude (Peylin et al., 2013), and its likely evolution in the future (e.g., Friedlingstein et al., 2006) all remain highly uncertain, contributing substantial uncertainty to the projections of future climate (Stocker et al., 2013). North American biogenic CO₂ fluxes, for example, are estimated on a 5-year, continentally aggregated basis to an accuracy no better than 50% (King et al., 2012; SOCCR, 2007). Individual annual estimates from biosphere models, biomass inventories, and atmospheric inversions (Hayes et al., 2012;
Peylin et al., 2013) also often diverge by a factor of 2. Uncertainties in CO₂ transport estimates may contribute a significant part of the flux uncertainties. With constraints from satellite column CO₂ observations, the flux uncertainties can be reduced (Crowell et al., 2019; Lauvaux et al., 2012; Liu et al., 2017).

Comprehensive measurements of regional atmospheric CO₂ distributions are urgently needed to develop a more complete understanding of CO₂ transport and sources and sinks. This is one of the key reasons NASA has been carrying out the Atmospheric Carbon and Transport–America (ACT–America) suborbital mission (Davis et al., 2017). Another ACT–America objective is to compare the full-column CO₂ spatial variability observed by the NASA Orbiting Carbon Observatory 2 (OCO-2) satellite mission with the partial-column CO₂ lidar measurements obtained below the ACT–America C-130 aircraft across the lower troposphere (Bell et al., 2020).

The target areas of the ACT–America mission are the mid-Atlantic, Midwest, and Gulf coast regions of the United States. The mission has completed four deployments with CO₂ lidar: one for each season (summer 2016; winter 2017; fall 2017; spring 2018) in each of these areas, as well as a fifth deployment without CO₂ lidar measuring summer a second time in 2019 due to the strong atmosphere-biosphere interaction during this season. Two NASA aircraft (C-130 and B-200) were deployed during each field campaign. Both aircraft were equipped with GPS for geographic location, altitude, and time information and in situ sensors for the measurement of atmospheric CO₂, other GHGs, and meteorological variables such as temperature, pressure, humidity, and wind. Besides these in situ sensors, the C-130 also carried remote sensors including the Multi-Functional Fiber Laser Lidar (MFLL; Dobler et al., 2013), which is the primary instrument for atmospheric column CO₂ measurements beneath the aircraft, and the Cloud Physics Lidar (CPL), an airborne backscatter lidar system designed specifically for studying clouds, aerosols, and atmospheric boundary layer (ABL) heights (McGill et al., 2002). The column CO₂ measurements are being used to evaluate OCO-2 XCO₂ observations (Bell et al., 2020), to understand XCO₂ distributions within typical midlatitude weather systems, and to evaluate CO₂ sinks and sources.

Potential XCO₂ measurement approaches include pulsed Integrated Path Differential Absorption (IPDA) (Abshire et al., 2013, 2018; Amediek et al., 2017; Refaat et al., 2015, 2020; Yu et al., 2017). A continuous-wave (CW) approach that uses heterodyne detection has also been investigated (Menzies et al., 2014; Spiers et al., 2011, 2016). Two intensity-modulated CW (IM-CW) approaches besides linear swept frequency use pseudo noise (PN) codes including direct PN modulation (Campbell et al., 2011; Quatrevalet et al., 2017) and BPSK (Campbell, Lin, Nehrir, Harrison, & Obland, 2014c). In addition, others have performed similar experiments using pulsed lidar to measure CO₂ in comparison with in situ sensors (Abshire et al., 2018; Amediek et al., 2017). MFLL is an IM-CW lidar using linear swept frequency modulation that was developed by the Harris, Corp. and the NASA Langley Research Center for column CO₂ measurements. The IPDA technique is used to measure column CO₂ differential absorption optical depth (DAOD) based on the combined lidar measurements at online and offline wavelengths using the CO₂ absorption line at 1.57111 μm (Browell et al., 2008; Dobler et al., 2013), and this corresponds to the peak center at 4,600-m altitude. The offlines were ±50 pm from the centerline. NASA Langley Research Center and the Harris, Corp. has jointly demonstrated its capability for a future space CO₂ lidar mission (Browell et al., 2009; Browell, Dobler, et al., 2010; Browell, Harrison, et al., 2010; Dobler et al., 2013; Lin et al., 2013). Previous test results from airborne technology demonstration flights have been published (Browell et al., 2009; Browell, Dobler, et al., 2010; Dobler et al., 2013; Lin et al., 2015). The signal-to-noise ratio (SNR) for clear sky IPDA measurements of CO₂ DAOD for a 10-s average over vegetated areas from a 7-km altitude was found to be as high as 1,300, resulting in an estimated precision uncertainty of 0.077% or an equivalent XCO₂ precision of ~0.3 ppm. A critical part of XCO₂ measurements is the ranging capability, which is realized by the use of orthogonal swept frequency (Campbell, 2013; Dobler et al., 2013) modulation techniques and super-resolution techniques (Campbell, Lin, Nehrir, Harrison, & Obland, 2014a, 2014b). The range uncertainty has been demonstrated to be under 1 m (Campbell, Lin, Nehrir, Harrison, & Obland, 2014a, 2014b). With this ranging capability, XCO₂ values can also be retrieved for the column between the aircraft and optically thick cloud decks and for weather conditions with intervening thin cirrus clouds or other aerosol layers (Lin et al., 2015).

The extensive ACT–America data set provides further opportunities for MFLL instrument evaluation and applications to carbon cycle science. Synoptic-scale variations in midlatitude atmospheric XCO₂ are on the order of one to several ppm (Bell et al., 2020; Chen et al., 2019; Pal et al., 2020), with variations...
occurring over length scales of a few to tens of kilometers; hence, MFLL measurements need to achieve precision of a few tenths of a ppm over distances of approximately 10 km and similar stability over the course of a flight. To reach this goal, dedicated MFLL calibration flights under clear to scattered cloud conditions were conducted and postflight data analyses were performed. This paper reports the specific MFLL data reduction algorithms used during ACT-America and the resulting DAOD and XCO2 retrievals. Generally, the data reduction covers various aspects of the airborne lidar data processing including consideration of lidar pointing geometry, data quality evaluation, cloudy/clear sky screening, and demodulation with a matched filter and uses of ancillary data such as aircraft time, location, and attitude records. The instrument and measurement approach are discussed in the next section. Experiments and calculation procedures associated with these data and data processing are also discussed. Section 3 contains calibration and MFLL measurements under different atmospheric conditions and an evaluation of MFLL measurement performance, including precision and accuracy during ACT-America. We conclude our main findings in section 4.

2. IM-CW Measurement Concept and MFLL Characteristics

2.1. Column CO2 Measurement Concept and Instrumentation

The CO2 lidar system, MFLL, deployed during four ACT-America field campaigns, has been extensively discussed previously (Browell et al., 2008, 2009; Browell, Dobler, et al., 2010; Browell, Harrison, et al., 2010; Campbell, Lin, & Nehrir, 2014; Campbell, Lin, Nehrir, Harrison, & Omland, 2014a, 2014b, 2014c; Dobler et al., 2013; Lin et al., 2013, 2015). Only a brief review is given here for the reader’s convenience. Detailed lidar system parameters can be found in Dobler et al. (2013). The MFLL system uses the IM-CW approach and is built with an all-fiber transmitter. It uses a 500-kHz modulation bandwidth, has a total laser output power of 5 W, and achieves common-mode noise rejection through the IPDA technique for all channels from many noise sources, including amplification, atmosphere, ground, and detector chain due to simultaneous transmission and reception of all the wavelengths. This differential absorption technique (or the ratio of lidar returns of two closely spaced wavelengths; c.f., Equation 2) removes all uncertainties caused by surface reflectivity and all other environmental conditions except CO2 and water vapor. The CO2 is what wanted, while the vapor differential absorption is corrected during retrieval (c.f., Equations 14 and 15).

The IM-CW method as implemented on MFLL encodes multiple wavelengths with orthogonal swept frequency amplitude modulation waveforms. The current instrument transmits three wavelengths (one online and two offline on both sides of online), but only two (online and the offline with shorter wavelength) are used in the processing. The signals are then combined before amplification and transmission to the atmosphere. The return signals are collected simultaneously on a single detector and digitized. The individual wavelength amplitudes are separated in the digital domain through correlation with a matched filter of the orthogonal swept frequency waveforms.

The concept of our IM-CW lidar system designed for CO2 IPDA measurements is illustrated in Figure 1. For the system described in this figure, two IM seed lasers with their distinct online and offline spectral properties, operating in conjunction with a near-infrared CO2 absorption line with one at the online absorption wavelength and the other at an offline absorption wavelength, are combined using fiber optics and simultaneously amplified by a single Erbium Doped Fiber Amplifier (EDFA) to increase the transmitted power. A small fraction of the transmitted beam is picked off via an optical tap inside of the EDFA and sent to a reference detector for power normalization.

This is necessary for accurate IPDA measurements because power fluctuations of the laser system would cause significant errors in XCO2 retrievals. The backscattered science signals of the online and offline
wavelengths from the surface as well as aerosols and clouds are simultaneously collected with a telescope, optically filtered with a narrow band optical filter, and detected by the science detector. Both the science and reference signals are amplified, electronically filtered, and then digitized for retrievals of column CO$_2$ using IPDA approach. Postprocessing of the digitized science and reference signals allows for discrimination between ground and intermediate scatterers using the matched filter technique. One also obtains differential absorption power ratios for inference of column CO$_2$ amounts as well as range estimates to the scattering targets.

In the type of CW lidar discussed here, a wide band (the bandwidth of the modulation is 500 kHz) modulation signal is used to modulate the intensity of a seed laser, which is then amplified and transmitted through the atmosphere (Figure 1). The modulation depth is approximately 90%. The digitized received signal is converted into a pulse through a mathematical transformation in the form of a correlation with a complex quadrature reference template stored in memory. The narrowness of the resulting pulse after the correlation is inversely proportional to the bandwidth of the modulation. Different types of modulation produce different pulse shapes or autocorrelation functions. One of the simplest modulations is linear swept frequency (chirp) and that is what is used here. For effective IM-CW performance, the modulation waveforms must exhibit mutual orthogonality (to avoid crosstalk in the demodulation process) and a long enough period to ensure that cloud returns and ground returns can be distinguished unambiguously. However, the type of optical amplifier used in MFLL’s transmitter only functions optimally when modulated above about 50 kHz. MFLL currently uses chirps whose instantaneous frequencies begin around 100 kHz and sweep up to about 600 kHz in 200 μs, a period that provides an unambiguous range of about 30 km. The number of sweeps per frame is 500, which provides a 10-Hz reporting rate of DAOD measurements. Each frame is processed by a matched filter using an FFT technique. With the 4-MHz signal sampling rate and long-swept-frequency waveforms, significant computational power is needed for both high-precision DAOD and high-resolution range retrievals. A super-resolution FFT interpolation technique is used in data processing. Details of creating this orthogonal modulation and others along with the super-resolution and interpolation techniques can be found in previous studies (Campbell, 2013; Campbell, Lin, & Nehrir, 2014; Campbell, Lin, Nehrir, Harrison, & Obland, 2014a, 2014b, 2014c). A convenient method for determining this correlation is to use

$$ R(\text{ref}, \text{data}) = \sum_{m=0}^{N-1} \text{ref}^*(m) \text{data}(m + n), \quad (1) $$

where $\text{ref}$ is the complex quadrature reference waveform and $\text{ref}^*$ is the complex conjugate of $\text{ref}$ (Campbell, 2013), $\text{data}$ are the data collected either from the reference or science detector, and $\text{DFT}$ is the digital Fourier transform. Once this is done on the transmit and receive channels, one may find the one-way DAOD by

$$ \tau_{\text{ meas}} = \frac{1}{2} \ln \left( \frac{P_{\text{off}}^P P_{\text{on}}^{*P}}{P_{\text{on}}^P P_{\text{off}}^{*P}} \right), \quad (2) $$

where $P_{\text{off}}^P$ is the received offline power, $P_{\text{on}}^P$ is the received online power, $P_{\text{off}}^{*P}$ is the transmitted offline power, and $P_{\text{on}}^{*P}$ is the transmitted online power.

Although the general architecture remains the same, significant changes to the MFLL system have been implemented since the publication by Dobler et al. (2013). Changes include the consolidation and synchronization of all computer functions, the separation of the single temperature-controlled chamber into three separately controlled chambers, and a microelectromechanical (MEMS) multiplexer to allow a Bristol 621A wavemeter to monitor all produced wavelengths. This has an accuracy of 0.2 pm, and these measurements are used to correct for wavelength drift in postprocessing. Additionally, we used a locked laser as a reference to track any potential changes in the wavemeter. For this mission, the MFLL with 5-W transmission power was installed in the NASA C-130 aircraft from the NASA Wallops Flight Facility. Its transmitter and telescope were pointed downward through a custom fused-silica window in the bottom of the fuselage. Note that significant degradation in the window coating was found during the winter 2017 field campaign.
resulting in a replacement of the window, which did not appear to have an impact on data calibration discussed in later sections.

The MFLL uses a simple custom telescope with a parabolic reflector coupled directly into a multimode fiber and housed in a carbon-fiber tube to limit thermal variability, and it is mounted with an approximate pitch offset of 3° relative to the aircraft to compensate for the average aircraft pitch during flight.

2.2. Instrument Zero-Path Calibration and Optical Depth Offset Subtraction

The MFLL calibration approach for compensation of the wavelength-dependent throughput of the internal optics utilizes a very near-field target with the assumption that the absorption over this very short path length is negligible and the power ratio of all channels should be unity after taking this zero-path calibration into consideration. In addition, this calibration accounts for differences in lidar signal path lengths within and outside the instrument such as differences in optical fiber length and the optical path from the telescope to the ground, which is important for correct range measurements, as well as correcting for optical or electrical frequency dependencies of the instrument that could impact the differential absorption measurement. We currently find the distance offset using the offline science channel distance minus the offline reference distance. That distance on the ground is the distance offset for zero path. We also measure the optical depth using Equation 2 on the ground that becomes our optical depth offset subtraction value. During the ACT-America campaigns, these short path tests were conducted during ground testing before each flight.

2.3. Ancillary Data for XCO2 Retrieval From DAOD

Atmospheric CO2 absorption is affected by the local meteorological state, including pressure, temperature, and humidity. Thus, meteorological conditions are needed in retrieving XCO2 from the MFLL DAOD measurements. During ACT-America flights, atmospheric CO2 and meteorological profiles were measured by in situ instruments on the C-130 and B-200 (Pal et al., 2020). In situ profiles measured during spiral segments of ACT-America flights were extremely useful for the calibration and validation of MFLL DAOD measurements; however, there were only a limited number of these profiles available. To calculate XCO2 values for all MFLL measured column CO2 DAODs throughout the campaign flights, meteorological profiles along the lidar measurement tracks were needed, as they are for the OCO-2 satellite measurements (O’Dell et al., 2012) and, of course, in situ data are not available at all locations and altitudes. For this study, Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2) reanalysis data (Bosilovich et al., 2017; Gelaro et al., 2017) were therefore used to derive XCO2. There are less than 30 model layers below the C-130 maximum flight altitudes. For the lidar XCO2 retrievals, third-order cubic spline interpolations are used, and the interpolating polynomials are integrated analytically to achieve the highest accuracy possible.

For the purpose of retrieving XCO2 values from the MFLL DAOD measurements at a specific location and time, atmospheric CO2 and water vapor absorption cross-sectional profiles were calculated at the MERRA-2 grid points and times based on their meteorological profiles including height, temperature, pressure, and humidity and with an assumed atmospheric constant CO2 mixing ratio (e.g., 400 ppm) and a spectroscopic model. The model estimated absorption cross-sectional profiles were then spatiotemporally interpolated using cubic spline to MFLL measurement track locations and times. These interpolated absorption profiles were then integrated over pressure from the surface to the aircraft altitude to obtain the model-equivalent optical depth for the 400-ppm XCO2 assumption. By scaling the MFLL measured CO2 DAOD with the modeled DAOD for XCO2 of 400 ppm, the MFLL-derived XCO2 value was retrieved. The model estimated water vapor DAOD, which is normally very small, was subtracted from the MFLL-measured DAOD to derive the measured CO2 DAOD before the scaling was done.

2.4. Range Measurements

The importance of ranging measurements in model DAOD and XCO2 retrievals was mentioned previously. The MFLL native vertical sampling resolution is 37.5 m, but through special techniques that take advantage of the high number of sweeps per frame of data (500 in this case), this resolution is improved by a factor of 500. This resolution improvement is accomplished using a special fine interpolation and super resolution technique that makes a transformation in the frequency domain of the matched filter correlation in real time (Campbell, Lin, Nehrir, Harrison, & Obland, 2014a). Once this is done and transformed back in to the spatial
domain, the peak location of the science and reference channels are recorded. To get the final range, the reference range is subtracted from the science range and calibration offsets are applied.

2.5. Attitude Correction

In retrieving the airborne measurements, certain data reduction procedures are applied. The first data screen is aircraft attitude. We only use data with pitch/roll angles less than 5°. This limits the off-nadir pointing to a maximum of ~750 m at the surface for a high-altitude flight at ~8.5 km. The retrieved nadir DAOD is estimated from the measured DAOD times a correction. This correction is estimated by finding the vertical component of the unit direction vector of the laser in the earth coordinate system. This is accomplished using an Euler angle transformation that takes into account the orientation of the MFLL with respect to the aircraft inertial navigation unit (INU), in addition to the rotation order of the INU.

2.6. Cloud and Measurement Screening

Besides aircraft attitude correction and range measurements, cloud detection and data screening are also performed. Low science signal levels are screened out as determined from the output of the matched filter and other parameters. Because we need to measure CO2 to the ground, we initially screen all thick and thin cloud cases from processing and use only clear sky cases for these results. Although it is technically possible to also include some thin cloud results, the SNR in those cases is significantly lower than for surface returns without interference from thin clouds or other scatterers, and we do not include them in the current analysis. Ranging capability discussed in previous sections is also critical for low cloud and ground discrimination. We currently distinguish between ground returns and cloud returns by comparing the distance we measured with known elevation data. If the difference in range to the surface is observed to be more than 100 m, it is assumed to be due to the intervening presence of a thick cloud. If the range comparison is within 100 m, it is assumed to be a ground return. The presence of multiple scatterers indicates the presence of thin clouds. Note that in our archived MFLL XCO2 data product (located at https://actamerica.ornl.gov/), we report these cloudy weather conditions with their corresponding cloud flags for cautions in using the data in these conditions.

2.7. Modeling Gas Absorption, Spectroscopy, and XCO2

2.7.1. Gas Absorption

As mentioned in previous sections, modeled DAOD values at an assumed gas concentration are needed in retrieving column gas concentrations from DAOD measurements. Optical depth for the kth species may be calculated from

$$\tau_k = \int_{h_{\text{surf}}}^{h_{\text{max}}} X_k DA \sigma_k dz,$$

where $X_k$ is the moist molar mixing ratio for the absorbing species, $D$ is the molar density, $A_p$ is Avogadro’s number, and $\sigma_k$ is the molecular absorption cross section for the kth species. By using the ideal gas law, the hydrostatic equation, and expressing the water vapor mixing ratio in terms of the specific humidity, it can be shown that

$$\tau_{\text{CO}_2} = \frac{A_p R_d}{\mathcal{R}} \int_{P_{\text{min}}}^{P_{\text{atm}}} \frac{X_{\text{CO}_2}'}{g} \frac{(1 - q_v)(\sigma_{\text{on}} - \sigma_{\text{off}})}{dP},$$

where $\tau_{\text{CO}_2}$ is the DAOD for CO2, $\sigma_{\text{on}}$ is the online molecular absorption cross section for the CO2 online wavelength, $\sigma_{\text{off}}$ is the online molecular absorption cross section for the CO2 offline wavelength, $R_d$ is the specific gas constant for dry air, $\mathcal{R}$ is the universal gas constant, $q_v$ is the specific humidity, $X_{\text{CO}_2}'$ is the dry molar mixing ratio for CO2, $g$ is the acceleration of gravity. For water vapor, the optical depth is given by

$$\tau_{\text{H}_2\text{O}} = \frac{A_p \mathcal{R}}{\mathcal{R}_w} \int_{P_{\text{min}}}^{P_{\text{atm}}} \frac{q_v}{g} (\sigma_{\text{on}}' - \sigma_{\text{off}}') dP,$$

where $\tau_{\text{H}_2\text{O}}$ is the DAOD for H2O and $\mathcal{R}_w$ is the specific gas constant for water vapor. The total DAOD in this model will be the sum of the two optical depths for CO2 and H2O, $\sigma_{\text{on}}'$ is the molecular absorption
cross section for water vapor online wavelength, and $\sigma_{\text{off}}$ is the molecular absorption cross section for water vapor offline wavelength. There may be a small insignificant contribution from other species at our wavelengths. This form is particularly useful because MERRA-2 reports specific humidity as one of the data products and pressure is the independent variable.

From the above, we see the dry column CO₂ must be

$$X_{\text{CO}_2} = \frac{A_v R_d P_{\text{top}}}{g} \int_{P_{\text{top}}}^{P_{\text{bottom}}} (1 - q_v(P))(\sigma_{\text{on}}(P) - \sigma_{\text{off}}(P))dP$$

$$= \frac{A_v R_d P_{\text{bottom}}}{g} \int_{P_{\text{bottom}}}^{P_{\text{top}}} (1 - q_v(P))(\sigma_{\text{on}}(P) - \sigma_{\text{off}}(P))dP$$

$$\approx \frac{W(P)}{\beta} \delta P,$$

where

$$W(P) = \frac{A_v R_d}{g} (1 - q_v(P))(\sigma_{\text{on}}(P) - \sigma_{\text{off}}(P))$$

is the dry air column weighting function. The uncertainty in CO₂ due to an uncertainty in pressure assuming a uniform mixing ratio is given by (Dufour & Bréon, 2003)

$$\frac{\delta X_{\text{CO}_2}}{X_{\text{CO}_2}} \approx \frac{W(P)}{\beta} \delta P = W(P) \frac{\delta P}{P},$$

where $W$ is the normalized weighting function. This is the absolute limit in error due to an error in pressure or range measurement assuming a perfect instrument and no other measurement errors. Near the surface $W(P) \approx 1$, so a 1-mb uncertainty in surface pressure leads to about a 0.4-ppm uncertainty in CO₂ for a single measurement. If the uncertainty is random, this may be improved further through averaging. A plot of the normalized weighting function is shown in Figure 2.

In this model, we need to take into account the variation of gravity with altitude and location. The acceleration of gravity at the surface may be calculated using the Somigliana Gravity Formula (van Dam et al., 2010)

$$g_0(\phi) = g_e \frac{1 + p \sin^2 \phi}{\sqrt{1 - e^2 \sin^2 \phi}}$$

where $\phi$ is the latitude, $p = 1.931851353 \times 10^{-3}$, $e^2 = 6.69438002290 \times 10^{-3}$, and $g_e = g_0(0) = 9.780318$ m/s². A height correction may be found by (Wenzel, 1989)

$$g - g_0(\phi) \approx (3.0877 \times 10^{-6} / s^2 - (4.3 \times 10^{-9} / s^2) \sin^2(\phi)) h + (7.2 \times 10^{-13} / ms^2) h^2.$$
evaluation of the 20012-0001 band at 2 μm. The ab initio intensities of the 30012-00001 band targeted here are systematically larger than those of Toth et al. (2006) by 2%. At the moment, it is still arguable if this change (in this particular band) is justified, as alternative ab initio calculations (Huang et al., 2014, 2017) favor Toth et al. (2006) data while independent experiments cannot provide conclusive justification (see for instance Devi et al., 1998; Jacquemart et al., 2012) at the required level of accuracy. New experiments and atmospheric validations are now underway to resolve this issue (c.f., Fleisher et al., 2019), and we may have to reevaluate our model to accommodate data from the official HITRAN2016 release.

2.7.3. Geopotential and Geometric Heights
Except for the calibration where meteorological vertical profiles can be derived from aircraft spiral measurements, all weather information along flight tracks is obtained from MERRA-2. The MERRA data contain profiles for temperature, pressure, geopotential height, and specific humidity, which are stored as four-dimensional arrays and are functions of time and location. It also includes surface pressure, sea level pressure, and geopotential surface height as a function of time and location. The hypsometric pressure and adiabatic temperature models are used to fill in missing profile data where necessary. All geopotential heights are converted to geometric heights. For geopotential height, a particularly useful closed form approximation discovered by M. J. Mahoney comes in the form where the geopotential height is given by (van Dam et al., 2010)

$$ Z \approx \frac{g_0(\phi)}{g_0(45^\circ)} \frac{h}{1 + h/R_e(\phi)} $$

where $$ Z $$ is the geopotential height and $$ h $$ is the geometric height

$$ R_e(\phi) = \frac{1}{\sqrt{\cos^2(\phi) a^2 + \sin^2(\phi) b^2}} $$

$$ a = R_e(0^\circ) = 6378137.0 \text{ m}, \text{ and } b = R_e(90^\circ) = 6356752.3 \text{ m}. $$ Solving this expression for geometric height gives

$$ h = \frac{g_0(45^\circ)}{g_0(\phi)} \frac{Z}{1 - (g_0(45^\circ)/g_0(\phi))Z/R_e(\phi)} $$

2.7.4. XCO₂ From DAOD Measurements
For simplicity, we model the measured DAOD as

$$ \tau_{\text{meas}} \approx \tau_{\text{CO₂}} + \tau_{H₂O} $$

To calculate XCO₂ from measured optical depth and MERRA-2 weather profiles, we therefore use

$$ \text{XCO₂} = \frac{\tau_{\text{meas}} - \tau_{H₂O}}{\tau_{\text{CO₂}}^\text{400 ppm}} $$

where $$ \tau_{\text{CO₂}}^\text{400 ppm} $$ is calculated assuming a constant 400 ppm in Equation 4.

2.8. Altitude-Dependent Bias Correction
Small differences were found as a function of range between modeled optical depths, as determined from Equations 4 and 5, and the in situ CO₂ mole fraction measurements, and MFLL measured optical depths, which are determined from Equation 2. There are multiple potential reasons causing these differences, such as the previously discussed uncertainties in the pressure-, temperature-, and humidity-dependent spectroscopic model, unknown cross talks between online and offline channels, nonlinear response of the instrument, and/or spatiotemporal differences between in situ profiling measurements and lidar overpasses. Even uncertainties in spectroscopic models could cause considerable differences (c.f., section 2.7.2). Another possibility is the shift of the absorption line near 1.5711 as a function of pressure. Such effects have been reported elsewhere (Ramanathan et al., 2013). As a result, for each ACT-America campaign, we
determined what the altitude-dependent bias correction needed to be to make the MFLL optical depths best agree with the model results. To determine the altitude bias correction, we collected all the available coincident lidar and in situ data. At multiple times and locations throughout each campaign, we collect MFLL observations over a cloud-free location where we also obtained an in situ profiles of CO₂ mole fraction from the altitude of the MFLL data collection to approximately 300 m above ground level (AGL) or lower in some cases where airport-missed approaches were arranged. The locations of all the profiles were reported in Pal (2019). All flights are conducted in relatively well-mixed, daytime conditions to minimize near-surface gradients. Because the retrieval is based on MERRA-2 meteorology, we then calculated the model optical depths from the MERRA-2 meteorological and in situ CO₂ profile measurements based on Equations 4 and 5. The advantage of using MERRA-2 in the calibration is whatever repeatable errors exist in MERRA-2, which can be calibrated out to a certain degree because the retrieval is based on the same meteorology. We then compared the model optical depths to the measured optical depths. Since we already subtract off the instrument optical depth and range calibration offsets from the ground testing, as discussed in section 2.3, we assume a relationship for the fractional change as

$$\frac{\tau_{\text{meas}} - \tau_{\text{mod}}}{\tau_{\text{meas}}} = k_1 + k_2 \tau_{\text{meas}}.$$  \hspace{1cm} (16)

To find $k_1$ and $k_2$, we minimize variance with respect to $k_1$ and $k_2$ which is given by

$$\sum_i \left( \frac{\tau_{\text{meas}, i} - \tau_{\text{mod}, i}}{\tau_{\text{meas}, i} - (k_1 + k_2 \tau_{\text{meas}, i})} \right)^2.$$  \hspace{1cm} (17)

Once $k_1$ and $k_2$ are found, the final correction is determined from

$$\tau' = \tau_{\text{meas}} - (k_1 + k_2 \tau_{\text{meas}}) \tau_{\text{meas}} = (1 - k_1) \tau_{\text{meas}} - k_2 \tau_{\text{meas}}^2.$$  \hspace{1cm} (18)

3. Results

3.1. Bias Correction

As an example, we take multiple altitude cases from spiral overpasses collected from the 2017–2018 campaigns where each altitude case represents one data point. The data points used are calculated from the left-hand side of Equation 16 by taking the difference between the measured optical depths and the modeled optical depths calculated from Equations 4 and 5 using MERRA-2 pressure, temperature, humidity, and in situ CO₂ measurements. In the case of CO₂, the airborne Picarro measurements were used. Once the in situ measurements were obtained, the modeled optical depths were calculated from our spectroscopy model. In this particular case, we found $k_1 = 0.01057$ and $k_2 = -0.04304$ (Spring 2018), $k_1 = 0.01035$ and $k_2 = -0.03979$ (Fall 2017), and $k_1 = 0.01063$ and $k_2 = -0.04114$ (Winter 2017). A plot of these results are shown in Figure 3a. The solid lines are the fits defined by Equation 16, and the dots are the data. Figure 3b was computed using $\Delta XCO₂ \approx 400\frac{\delta r}{\tau_{\text{meas}}}$, where $\delta r$ is the difference between the measured points and the straight line fits. Conversely, we can estimate the maximum change due to the bias correction as $\Delta XCO₂ \approx 400(k_1 + k_2 \tau_{\text{max}})$ in each case, the maximum change is approximately 4 ppm or less. The maximum difference between each season can be computed by finding the difference of the maximum change for each season. This is about 0.5 ppm.

A plot of the residual XCO₂ errors in ppm are shown in Figure 3b. In this case, the mean is zero with a standard deviation of 0.8 ppm. When the aircraft was closer to the ground, atmospheric gas absorption would be smaller. Thus, the lidar absorption signals would be weak, and retrieved DAOD values as shown in Figure 3b would be noisier than for higher DAODs. Once the bias calibration of Equation 18 is applied to MFLL measurements, the final XCO₂ is estimated from the scaling of measured CO₂ DAOD and model DAOD based on meteorological state profile data with assumed 400-ppm CO₂ (c.f., Equation 15). The most encouraging result is the consistency between altitude-dependent calibration results going from season to season and in different measurement environments. This consistency gives some indication of the long-term stability of the instrument and shows a potential that if it is calibrated in one field campaign, then the calibration...
more or less holds for future measurements. By calculating the difference between the straight line fits in Figure 3, we found the maximum equivalent difference between each calibration is less than 0.5 ppm.

### 3.2. In-Flight Testing of Measurement Precision and Stability

To fully understand instrument performance, besides absolute accuracy, other key parameters such as measurement precision, as reflected by measurement SNR, and in-flight instrument stability over hours need to be evaluated. Before discussing this, it should be pointed out that this topic has been addressed in a previous study (Lin et al., 2013), where specific instrument parameters were modeled and precision and accuracy were calculated. For this study, dedicated MFLL flight experiments were conducted. The selected region was the Gulf of Mexico, which was chosen due to the relatively homogeneous conditions there during onshore flow. On 6 November 2017, the synoptic set-up over the southern portion of the Gulf States and over the Gulf of Mexico was characterized with a broad high-pressure system with moderate southerly wind in the boundary layer. The flight plans for both C-130 and B-200 aircraft (Figure 4a) were designed to examine the performance of the MFLL system under homogeneous atmospheric condition over water. The B-200 aircraft made two west-east ABL legs at an altitude of 400 m above mean sea level (MSL), while the C-130 made five west-east transacts in the free troposphere (FT) at an altitude of 4,800 m above MSL; additionally, five vertical profiles were made over water using both aircraft which confirmed a spatially homogeneous boundary layer depth of 700 m above MSL over water (not shown here).

Figure 4b displays a longitude-versus-altitude cross section of the in situ measurements of atmospheric CO2. The column averaged CO2 mole fraction obtained from in situ measurements was 404.0 ppm. In Figure 4b, two ABL legs are stacked on each other (same latitude); thus, we separated the two ABL legs in Figure 4c where we present a time series view of CO2 variability from west-to-east within the ABL. These data confirm that CO2 distributions along ABL-Leg 1 and ABL-Leg 2 were similar; thus, atmospheric CO2 was steady with respect to time since ABL-Leg 2 was flown more than an hour after ABL-Leg 1. Figure 4c illustrates that the CO2 horizontal distribution in the boundary layer over the water was also spatially homogeneous during both transacts. The mean, median, and standard deviation of CO2 measurements along ABL-Leg 1 (ABL-Leg 2) were 405.15 (405.11 ppm), 405.17 (405.29 ppm), and 0.09 ppm (0.08 ppm), respectively. These in situ observations demonstrate that although there was some vertical variation, the CO2 variability at sampled constant altitudes was minimal, even within the ABL. Thus, spatiotemporal XCO2 variations must be small for this case, which is what was desired for our precision and stability testing.

Figure 5 shows the 1-, 10-, and 60-s running mean variations of the XCO2 retrieved from the MFLL measurements. The native instrument measurement rate is 10 Hz, and the values of XCO2 retrievals at this 10-Hz reporting rate is dominated by instrument noise and needed to be further averaged before science use. Although the CO2 lidar system maintained continuous operation during the entire flight period, the plotted data showed some gaps that resulted from removing retrievals during aircraft turns and from cloudy scenes.
It can be seen that with the longer averaging times, the random errors reduced significantly. Generally, when the averaging time was longer than 10 s, the variation in retrieved XCO$_2$ was within 1 ppm.

The estimated precision (standard deviation) for 0.1-, 1-, 10-, and 60-s averages were determined to be 3.4, 1.2, 0.43, and 0.26 ppm, respectively. Their corresponding SNRs, which are the mean XCO$_2$ divided by the standard deviation, were found to be 120, 330, 950, and 1,600, respectively. The 10-s results are very close

**Figure 4.** (a) Ground tracks of B-200 (blue) and C-130 (red) aircraft on 6 November 2017 during ACT-America fall field campaign. (b) Longitude-versus-altitude cross section of 5-s averaged samples of CO$_2$ (ppm) obtained by Picarro onboard both C-130 and B-200. (c) Time series view of east-west transacts of CO$_2$ in the boundary layer over Gulf of Mexico along two identical tracks: Leg 1 (upper panel) and Leg 2 (lower panel) (c).

It can be seen that with the longer averaging times, the random errors reduced significantly. Generally, when the averaging time was longer than 10 s, the variation in retrieved XCO$_2$ was within 1 ppm.

The estimated precision (standard deviation) for 0.1-, 1-, 10-, and 60-s averages were determined to be 3.4, 1.2, 0.43, and 0.26 ppm, respectively. Their corresponding SNRs, which are the mean XCO$_2$ divided by the standard deviation, were found to be 120, 330, 950, and 1,600, respectively. The 10-s results are very close

**Figure 5.** Variations of the retrieved XCO$_2$ for the Gulf of Mexico flight on 6 November 2017 for the repeated passes discussed in the text and shown in Figure 3. The CO$_2$ variation as measured by Picarro was less than 0.1 ppm, thus making this segment suitable for stability testing. Here, the 1-, 10-, and 60-s running means, as indicated by their corresponding color, are shown. The flight path for this data covers approximately 700 km.
to previously reported MFLL results (Dobler et al., 2013) after surface type and flight altitude differences are taken into consideration and consistent with that predicted in a previous study (Lin et al., 2013).

Column XCO₂ calculated from MFLL (Figure 5) shows very little change during the five repeated passes over this portion of the Gulf of Mexico, consistent with the homogeneous, steady-state atmospheric conditions described in Figure 4. For the in-flight instrument stability, the trends at 20-min, 30-min, and 1-hr scales in the 10-s averaged Gulf data were analyzed. No statistically significant drifts were found. On the 1-hr scale, the drift is below 0.1 ppm.

This flight duration over the Gulf of Mexico was generally very similar to that of OCO-2 under flights. However, OCO-2 under flights were conducted at about 8.5 km above MSL altitudes over land. Since land surface lidar reflectance is about 4 times greater than over water (Dobler et al., 2013), lidar return signals from the sea surface at this 4.8 km altitude would be weaker than those for OCO-2 under flights. Other ACT-America flights were equal to or lower than those of OCO-2 under flights, and nearly all were over land. Thus, lidar signal level from this experiment would be among the weakest for ACT-America flight campaigns. The estimated precision and SNR would be a conservative result for normal ACT-America instrument operations.

We additionally evaluate instrument precision from an 8.4-km altitude OCO-2 under flight conducted across the US Great Plains on 22 October 2017. The flight track and MFLL observed XCO₂ are shown in Figures 6a and 6b, respectively. In this case, some of the observed XCO₂ variations were most likely caused by atmospheric differences in CO₂ concentrations, variability in meteorological state variables, and boundary layer thickness and surface height variations. The estimated precision for 0.1-, 1-, 10-, and 60-s averages was calculated as 4.6, 1.5, 0.82, and 0.46 ppm, respectively. Their corresponding SNRs were 88, 270, 490, and 909, respectively.
respectively. Note that the instrument stability in this case should stay the same as that of oceanic case since the environment conditions such as temperature of the instrument were the same and well controlled. To facilitate the validation of remote-sensing measurements with in situ data, NASA’s Global Modeling and Assimilation Office (GMAO) assimilates the in situ data into the Goddard Earth Observing System (GEOS). The result, a “gap-filled,” two-dimensional, along-track cross section, referred to here as a curtain, is then directly comparable to the remote-sensing measurements taken on the same platform. To produce the curtains, the assimilation system uses a model configuration similar to that described in Ott et al. (2015) and an analysis methodology derived from that used to produce the three-dimensional fields of OCO-2 data highlighted in Eldering et al. (2017). For more details about the differences from the system described in those publications, see Bell et al. (2020). The general consistency of these MFLL XCO2 measurements when compared to GEOS assimilated results and OCO-2 observations (not shown here, c.f., Bell et al., 2020) further demonstrates the stability and precision of the MFLL XCO2 measurements.

For both the Gulf and land cases and with 10-s and longer averaging times, the MFLL XCO2 measurement precision clearly meets the ACT-America science requirement, which is 1 ppm at 20-km spatial scale corresponding to about 2.6 min of C-130 flight time. Even the maximum deviation of about 0.8 ppm, as shown in the green curve in Figure 4 for 60-s averaged data, is well below 1 ppm.

4. Conclusion

We have demonstrated a technique for measuring XCO2 with an IM-CW lidar system through flight testing with very good results. The lidar measurement precision for 1-, 10-, and 60-s averages (approximately 0.13, 1.3, and 8 km horizontally) is between 0.9–1.2, 0.4–0.6, and 0.2–0.4 ppm, respectively, and on 1-hr time scales, the average drift is below 0.1 ppm, which is small compared to typical atmospheric variations in atmospheric CO2. These results demonstrate that the precision and stability of our CO2 lidar measurements meet ACT-America science requirements. Furthermore, our technique is suitable as a standard tool for remotely measuring atmospheric CO2 column concentrations with an airborne lidar system that has been in development and testing for many years. We found our altitude-dependent calibration holds over all four deployments in four seasons which means that we can calibrate it using in situ profiling observations as a reference in one field campaign and have reasonable expectation the same calibration applies for future campaigns without the need for extensive recalibration flight experiments. We found that our XCO2 measurement accuracy was about 0.8 ppm compared against our model results derived from in situ CO2 measurements based on the state-of-the-art HITRAN spectroscopic model and MERRA-2 meteorology. By using meteorology consistent with the retrieval in the calibration, we can take out potential bias errors in the derived XCO2 retrieval caused by bias errors in MERRA-2. There is still an unanswered question regarding the cause(s) of the altitude-dependent bias errors even though we have demonstrated that we can calibrate it for it. In the future, we plan to investigate this question and apply the MFLL XCO2 measurements in studying CO2 variability in different atmospheric conditions being investigated in ACT-America.

Data Availability Statement

ACT-America data supporting this work can be found online (https://www-air.larc.nasa.gov/missions/ACT-America/ and https://doi.org/10.3334/ORNLDAAC/1649). MERRA-2 meteorological data can be found online (https://gmao.gsfc.nasa.gov/reanalysis/MERRA-2/data_access/).

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