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Low altitude CO_2 from the Atmospheric Chemistry Experiment (ACE) satellite

Peter Bernath[a,b,c,*], Chris Boone[b], Anton Fernando[c], Scott Jones[b]

[a] Department of Chemistry and Biochemistry, Old Dominion University, Norfolk, VA 23529, USA
[b] Department of Chemistry, University of Waterloo, Waterloo, ON N2L 3G1, Canada
[c] Department of Physics, Old Dominion University, Norfolk, VA 23529, USA

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ABSTRACT

The Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) has been measuring atmospheric composition by solar occultation from low Earth orbit since 2004. A new version (4.0) of ACE-FTS processing has added low altitude CO_2 as a routine data product. ACE provides a near global data set of altitude profiles of CO_2 volume mixing ratios (VMRs) on a 1 km grid from 5 to 18 km. We provide an initial evaluation of these data using occultations from the month of May for 2004–2017 in the 55°–70°S latitude range. Comparisons are made with ground-based measurements at Macquarie Island, the South Pole, the CarbonTracker 2017 model and G. Toon's empirical model. Agreement for trends is good, but ACE-FTS VMRs have a low bias at 5.5 and 6.5 km in altitude.

1. Introduction

CO_2 is the most important anthropogenic greenhouse gas in the atmosphere [1]. As the main non-condensable greenhouse gas, it functions as the "climate control nob" for the Earth [2]. Understanding the sources and sinks of carbon dioxide is therefore essential for modeling and predicting climate change.

Numerous measurements of atmospheric CO_2 have improved our knowledge of the carbon cycle and the global carbon budget [3]. For example, regular CO_2 observations by NOAA (National Oceanic and Atmospheric Administration) include the analysis of flask samples and continuous *in situ* measurements at remote sites such as Mauna Loa, Hawaii (https://www.esrl.noaa.gov/gmd/ccgg/). Altitude information is provided by tall towers and measurements by small aircraft typically sampling up to 8 km [4]. Commercial aircraft can fly higher; the CONTRAIL (Comprehensive Observation Network for TRace gases by AIrLiner) program [5] and CARIBIC (Civil Aircraft for Regular Investigation of the Atmosphere Based on an Instrument Container) system [6] extend sampling up to about 12 km. Dedicated aircraft campaigns can provide altitude-resolved CO_2 concentrations with a wide geographic coverage [e.g., 7] and high-altitude aircraft such as the ER-2 can measure up to about 21 km [8]. High altitude balloons, using for example the AirCore sampling system [9], can measure up to 30 km.

CO_2 is also measured by remote sensing from the ground and from orbit. The total atmospheric column of CO_2 is measured by ground-based Fourier transform infrared absorption spectroscopy using the Sun as a light source by the TCCON (Total Carbon Column Observing Network) network [10]. Total column measurements are also being made from orbit using reflected/scattered sunlight in the near infrared by the OCO-2 (Orbiting Carbon Observatory-2) grating spectrograph [11] and the GOSAT (Greenhouse gases Observing SATellite) Fourier transform spectrometer (FTS) [12].

CO_2 observations in the thermal infrared available from TES (Tropospheric Emission Spectrometer) on the Aura satellite (to 2018) [13], AIRS on the Aqua satellite [14] and IASI (Infrared Atmospheric Sounding Interferometer) on MetOP [15] sample mainly the mid-troposphere. In addition, limb sounding instruments such as ACE-FTS (Atmospheric Chemistry Experiment Fourier Transform Spectrometer) [16], MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) and SABER (Sounding of the Atmosphere using Broadband Emission Radiometry) [17] provide a high altitude (mesosphere and lower thermosphere) altitude-resolved CO_2 data product.

These CO_2 measurements are used for a variety of scientific studies. As a long-lived molecule, CO_2 is an excellent tracer of atmospheric dynamics and has been used to determine the stratospheric age-of-air [e.g., 18] and to evaluate the accuracy of transport in models [4,19]. NOAA observations have been assimilated

* Corresponding author at: Department of Chemistry and Biochemistry, Old Dominion University, 4541 Hampton Blvd, Norfolk, VA 23529, USA.
E-mail address: pbernath@odu.edu (P. Bernath).

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into the CarbonTracker (CT2017; https://www.esrl.noaa.gov/gmd/ccgg/carbontracker/) model that predicts CO_2 as a function of latitude, longitude, altitude and time [20]. Perhaps the most important application is "inverse" modeling to determine the sources and sinks of CO_2 to improve our understanding of the carbon cycle. For example, a recent paper by Crowell et al. [21] reports on global and regional flux inversions from 9 different modeling groups based on OCO-2 total column CO_2 measurements. These "top-down" estimates yield a total global carbon sink of 3.7 ± 0.5 PgC per year and a total land sink of 1.5 ± 0.6 PgC per year for 2015–2016.

Missing from all of these data sets are global altitude-resolved CO_2 measurements in the upper troposphere and lower stratosphere. These data are now available for the 5–18 km altitude range from version 4.0 of ACE-FTS processing for 2004 to the present. Low altitude ACE CO_2 can be assimilated by models or used to validate global models such as CarbonTracker, and to improve CO_2 flux inversions [22].

2. Methods

The new long-term (currently 14 years) CO_2 data set is from the Atmospheric Chemistry Experiment Fourier Transform Spectrometer (ACE-FTS) on the SCISAT satellite. ACE provides a set of simultaneous measurements of trace gases, thin clouds, aerosols and temperature using solar occultation from a small satellite [23]. The circular low-earth orbit (650 km, 74° inclination to the equator) gives coverage of tropical, mid-latitude and polar regions (see Fig. 6 in reference 23) with particularly good coverage at high latitudes. The primary instrument is a high resolution (0.02 cm^{-1}) infrared FTS operating from 2 to 13 μm (750–4400 cm^{-1}). During sunrise and sunset, the FTS measures an atmospheric absorption spectrum in the limb viewing geometry every 2 s. An occultation therefore consists of typically 30–100 spectra (depending on the geometry) with different tangent heights (vertical distance from the light ray to the ground) and slant paths. These spectra are analyzed on the ground at the University of Waterloo (see http://www.ace.uwaterloo.ca/) to obtain vertical concentration profiles of atmospheric constituents. The nominal vertical resolution of ACE-FTS, as set by the circular field of view of 1.25 mrad is about 4 km projected on the limb, but the effective resolution is determined by the vertical sampling and is about 2 km from the cloud tops (>5 km) in the upper troposphere/lower stratosphere. A single ray in the center of the field of view is used for ray tracing.

The new version of ACE-FTS processing (v.4.0) has made many improvements over the previous version (v.3.5/3.6) as described elsewhere [24,25, and in preparation] and briefly below for low altitudes. This paper is the third in a series on v.4.0 that starts with a description of a new lineshape function [24], followed by the use of the nitrogen continuum to obtain pointing (i.e., tangent heights) at low altitudes [25]. Low altitude temperature and pressure are from the Canadian weather model [25]. ACE processing models the atmosphere with a 1 km altitude grid to 150 km beginning at 0.5 km from the ground. The forward model calculates the atmospheric transmission of the solar ray to match the observed spectrum using primarily the HITRAN 2016 [26] line parameters and absorption cross sections, along with a new empirical instrument lineshape function [24].

The first step in data processing is to determine the temperature and pressure in each 1 km layer and the pointing (i.e., tangent heights) of the measurements. Below 18 km, temperature (T(z)) and pressure (P(z)) are obtained from the analysis run of the global Canadian weather service model [27] and pointing is obtained using the collision-induced absorption (CIA) of N_2 near 2500 cm^{-1} [25]. Boone and Bernath [25] discuss this method in detail and provide lists of "microwindows" (short segments of spectra) and interfering molecules used for N_2–CIA analysis and for low altitude CO_2 retrievals. The CO_2 "reference standards" are the spectroscopic parameters as given in HITRAN 2016 [26] for the set of CO_2 lines [25] used in the retrieval. As discussed by Boone and Bernath [25], v4.0 "operational" CO_2 retrievals improve on previous preliminary work [e.g., 28,29].

Briefly, the microwindows employed in the low altitude CO_2 retrieval range from 3160 to 3380 cm^{-1}, avoiding the region near 3250 cm^{-1} where severe signal losses were experienced early in the mission from ice build-up on the detector windows. The continuum signal to noise ratio in this wavenumber region is between 100:1 and 150:1. The CO_2 lines in these microwindows were selected because they do not saturate in ACE-FTS measurements, and they exhibit relatively low sensitivity to temperature. V.4.0 analysis uses a much more accurate (compared to previous ACE-FTS processing versions) *a priori* empirical description of CO_2 profiles in the troposphere and stratosphere as provided by G. Toon. His CO_2 model is currently used to generate *a priori* CO_2 profiles for the TCCON network of ground-based Fourier transform spectrometers [10] used to validate the NASA OCO-2 mission. Volume mixing ratio (VMR) profiles generated from Toon's CO_2 model are also used to determine ACE-FTS pointing above 18 km to about 60 km. Because CO_2 is fixed to an assumed profile in this altitude region when generating tangent heights, it is not possible to perform an independent retrieval of CO_2 between 18 and 60 km.

3. Results and discussion

V.4.0 started by processing occultations for the month of May for the period 2004–2017, which provided a data set for assessing data quality. The occultations in the Southern Hemisphere span a relatively compact range of latitudes, as shown in Fig. 1 using 2° latitude bins. This narrow geographic focus, repeating from year to year, in a time period absent the disruptive influence of a polar vortex (which forms during winter), presents an opportunity for preliminary trend studies. For an initial analysis of the new CO_2 product, we focus on the 4408 occultations in the 55°–70°S latitude region from the fourteen months of data (i.e., the May data spanning fourteen years). Because of the ACE orbit, Northern Hemisphere occultations are distributed over a much wider geographic region for the month of May and are not considered further. ACE-FTS v.4.0 low altitude CO_2 VMR profiles (from 5 to 18 km) are reported on an irregular altitude grid (defined by the measurement tangent heights) and a standard 1 km altitude grid. The results presented here are average profiles from data on the 1 km grid.

ACE-FTS data sometime contain unphysical values, for example when data losses during downlink from the satellite yield large altitude gaps in the measurements for a particular occultation, necessitating a filtering of the data set. Large positive and negative VMR values were removed and then VMRs more than 2 standard deviations away from the mission average at each altitude level were discarded. In all, 4% of the data were removed.

Fig. 2 plots the average altitude profiles of CO_2 VMRs for each May for 2004–2017, 55°–70°S. The average profile for May 2017 with one standard deviation errors is displayed in Fig. 3, along with the average temperature profile. The standard deviation at each altitude of these CO_2 profiles is about 1.5% (~6 ppm) while the typical statistical error estimated from the random fitting error in the least squares analysis for an individual measurement is about 2.5% (~10 ppm). For comparison with Fig. 2, the corresponding *a priori* values from Toon's empirical model are provided in Fig. 4. The *a priori* values were calculated for each occultation used and averaged for the month of May. (Note that ACE-FTS processing does not use optimal estimation so these *a priori* values serve only as initial guesses for the retrievals.)

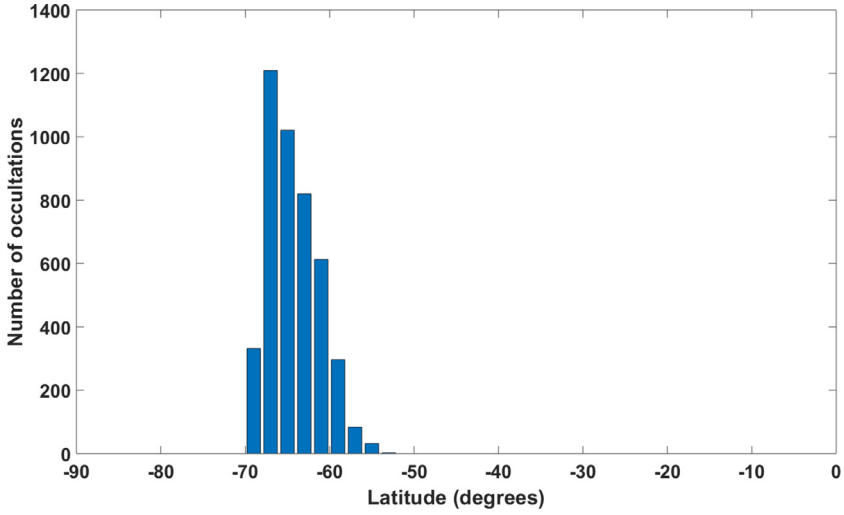

Fig. 1. Latitude distribution of May occultations in the Southern Hemisphere for 2004–2017.

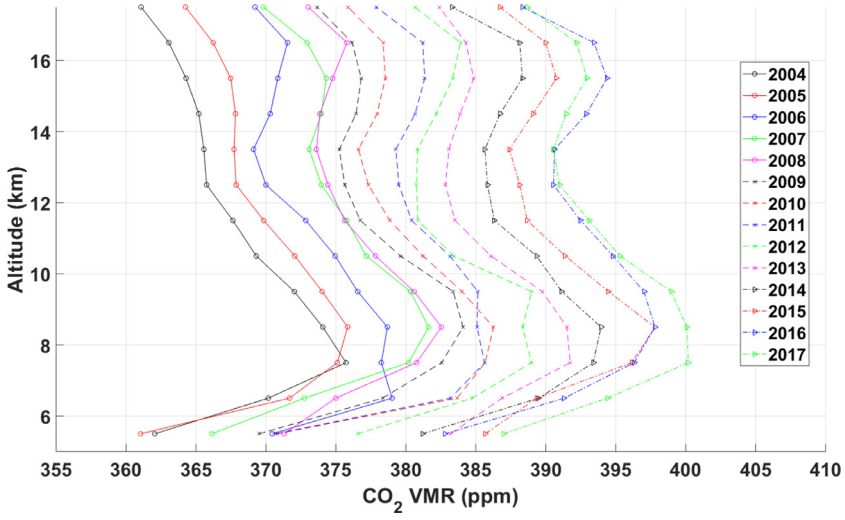

Fig. 2. Average altitude profiles for CO_2 VMRs for each May from 2004 to 2017 for 55°–70°S.

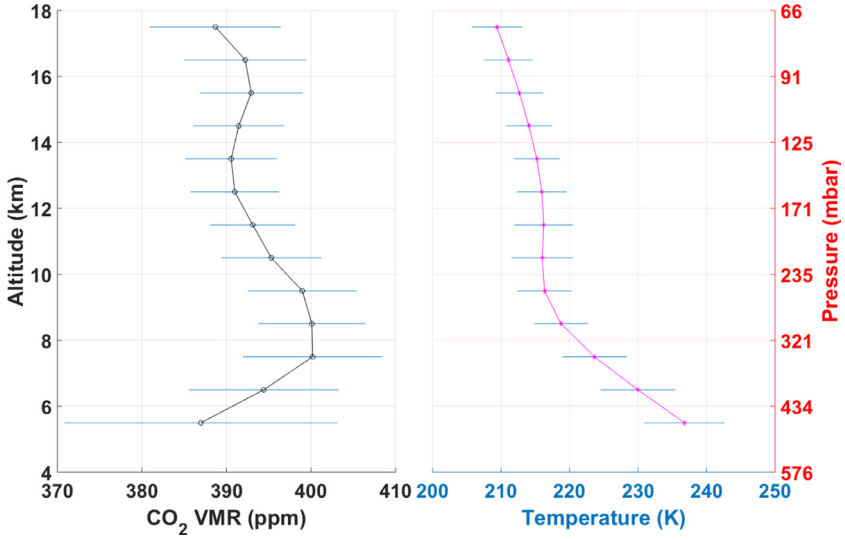

Fig. 3. CO_2 altitude profile (left panel) and temperature (right panel) for May 2017 with one standard deviation error bars for the 55°–70°S region. The average atmospheric pressure is given on the far right in mbar (hPa).

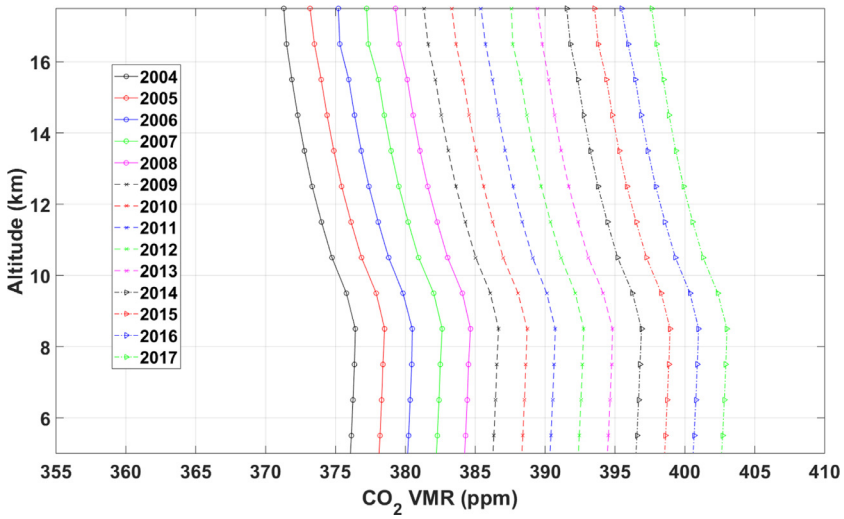

Fig. 4. CO_2 altitude profiles for each May 2004–2017 for 55°–70°S from Toon's empirical model, assuming a 9 km tropopause altitude.

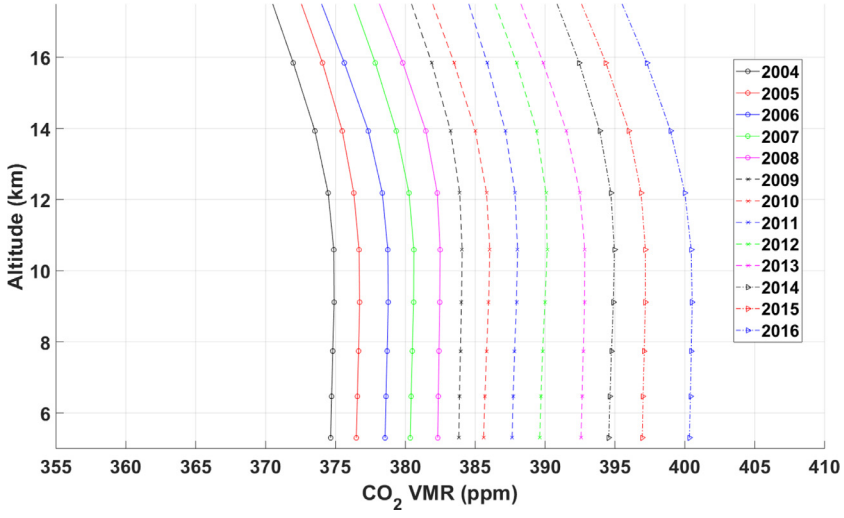

Fig. 5. CO_2 altitude profiles from CarbonTracker 2017.

CarbonTracker [20] (CT2017; http://carbontracker.noaa.gov) is a sophisticated global CO_2 model that has assimilated observations made at hundreds of sites around the world by more than 50 laboratories. NOAA provides these CO_2 measurements as the GLOBALVIEW+ data product (available from NOAA's ObsPack web site https://www.esrl.noaa.gov/gmd/ccgg/obspack/). In addition to surface sites, GLOBALVIEW+ includes data from tall towers, aircraft campaigns, shipboard measurements, CONTRAIL [5] and AirCore [9]. CT2017 has predictions at altitudes of 5.3 km, 6.5 km, 7.7 km, 9.1 km, 10.6 km, 12.2 km, 13.9 km and 15.8 km within the 5–18 km range of ACE-FTS low altitude CO_2 data. CO_2 VMR values for the month of May (2004–2016) were obtained from the CT2017 web site (http://carbontracker.noaa.gov). VMR averages were calculated for 9 altitude levels (in the 5.3 km to 18 km range) for all the longitudes and the latitudes between 55°S and 69°S. These profiles are plotted in Fig. 5.

In general, the agreement for CO_2 VMRs as depicted in Figs. 2, 4 and 5 is reasonable considering the ACE-FTS error bars (Fig. 3). The tropopause height for 55°–70°S is about 9.5 km (Fig. 3) and so CO_2 should be well mixed below this altitude. The retrieved CO_2 VMR values at 5.5 and 6.5 km in Fig. 2 exhibit a clear low bias compared to expectations from the models (Figs. 4 and 5). This could trace back to difficulties in the determination of tangent heights from the N_2 continuum at these altitudes [25], difficulties in the CO_2 VMR retrieval in this altitude region from the chosen set of microwindows [25], or a combination of the two effects. ACE VMRs in the stratosphere also decrease more rapidly than CT2017 and Toon's model (Figs. 2, 4 and 5). There is also a peak in the CO_2 profile near 15.5 km (Fig. 3), which may be an artifact. The behavior of VMR profiles for other molecules in this altitude region should be evaluated to provide further insight. As will be demonstrated below, although these data are biased, they yield reliable trend values, as is often the case for ACE-FTS data.

The ACE-FTS values in the troposphere should match ground-based measurements. There are relatively few CO_2 measurements in the Southern Hemisphere, but high-quality ground-based data are available for Macquarie Island [30]. Macquarie Island is a small (34 km long and 5 km wide) uninhabited island located between New Zealand and Antarctica at 54°37′ S, 158°52′ E that falls near the 55°–70°S latitude range considered here. From 2005 to 2016, Australia's Commonwealth Scientific and Industrial Research Organisation (CSIRO) operated a well calibrated non-dispersive infrared spectrometer that continuously measured CO_2 VMRs on Macquarie Island with a sampling inlet about 13 m above sea level. Estimated CO_2 VMR values were extracted from the time series

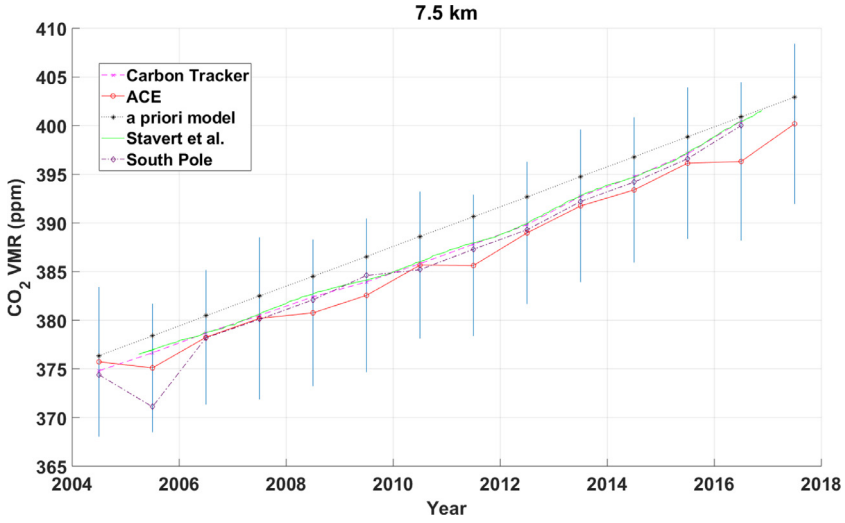

Fig. 6. Trend comparisons at 7.5 km.

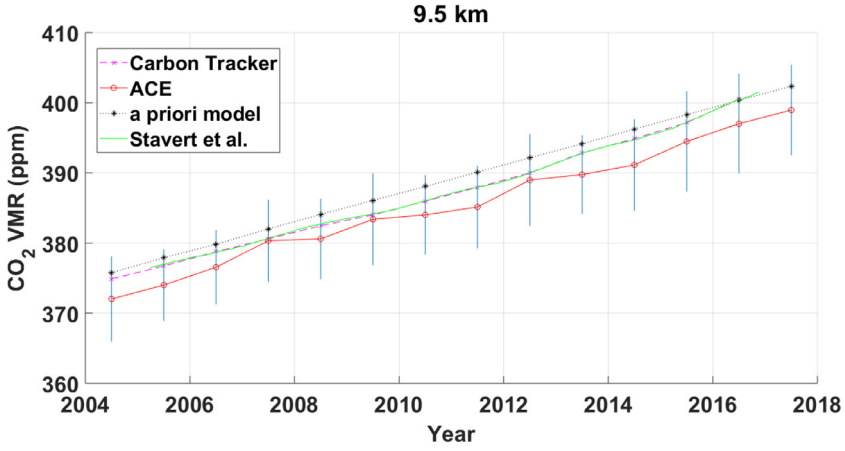

Fig. 7. Trend comparisons at 9.5 km.

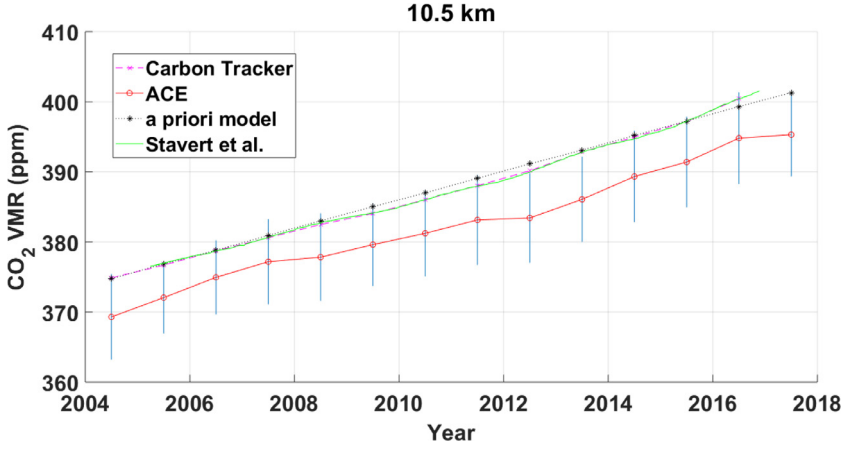

Fig. 8. Trend comparisons at 10.5 km.

in Fig. 10a of Stavert et al. [30] because numerical data were not available.

Ground-based flask measurements are also made by NOAA at the South Pole at an altitude of 2.84 km. P. Tans (private communication) provided CO_2 VMR data for each May from 2005 to 2016.

The ACE-FTS CO_2 time series for the 13 altitudes between 5 and 18 km on the 1 km grid were obtained and the VMRs with one standard deviation errors are available as supplementary data. The time series were fitted by weighted linear regression and trend values for 2004–2017 are given in Table 1. The CO_2 trends in Table 1 seem to increase with altitude (although the statistical error bars are substantial, and no systemic errors are estimated) even though the upper altitudes in the stratosphere have older air which has a smaller trend value (https://www.esrl.noaa.gov/gmd/ccgg/trends/). ACE CO_2 retrievals depend on the accuracy of the temperature and pressure (and thus the

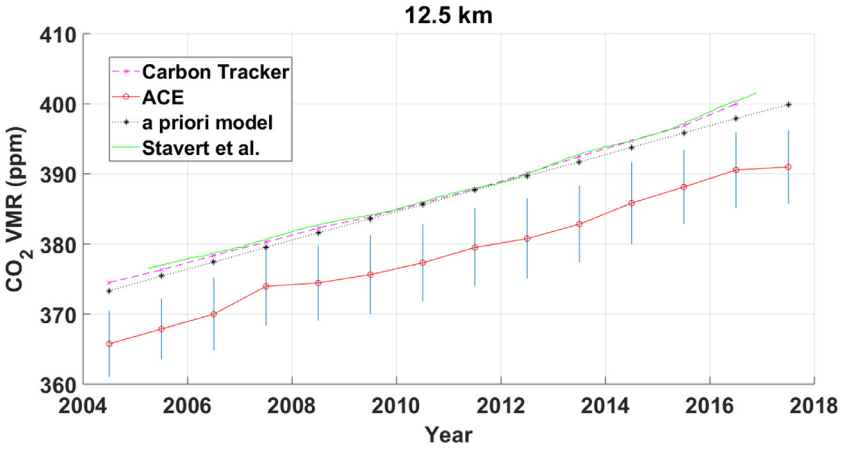

Fig. 9. Trend comparisons at 12.5 km.

Table 1
Linear trends of ACE-FTS CO_2 for altitude levels.

Altitude (km)	Linear trend (ppm/year)	σ (ppm/year)
5.5	1.9	0.2
6.5	1.8	0.1
7.5	1.93	0.07
8.5	1.96	0.08
9.5	1.98	0.06
10.5	1.91	0.06
11.5	1.89	0.08
12.5	1.95	0.05
13.5	1.97	0.05
14.5	2.09	0.06
15.5	2.27	0.07
16.5	2.23	0.09
17.5	2.13	0.08

tropopause height) from the Canadian weather service model; any errors in these values result in systematic errors in our retrievals.

In order to match CT2017 altitudes (7.7 km, 9.1 km, 10.6 km, 13.9 km) trend plots were made for ACE-FTS altitudes of 7.5 km, 9.5 km, 10.5 km and 12.5 km, respectively (Figs. 6–9). One standard deviation error bars are included for the ACE data. Toon's *a priori* model data, the values at the South Pole and data obtained on Macquarie Island by Stavert et al. [30] are also plotted. The linear trend for the Macquarie Island time series (2004 - 2017) is 2.08 ± 0.02 ppm/year, the South Pole linear trend for 2005–2016 is 2.2 ± 0.1 ppm/year, the trend of the *a priori* model time series (2004–2017) is 2.04 ± 0.05 ppm/year for the altitudes 7.5 km–17.5 km and the trend for the CT2017 time series (2004–2016) is 2.06 ± 0.05 ppm/year for the altitudes 5.3 km, 6.5 km, 7.7 km, 9.1 km, 10.6 km, 12.2 km, 13.9 km 15.8 km and 18 km. Within two standard deviations the ACE-FTS trend values in Table 1 are in general agreement with the values for Macquarie Island, the South Pole, Toon's model and CT2017.

As already mentioned, the ACE-FTS CO_2 VMRs decrease more rapidly with altitude than both CT2017 and Toon's model, as is evident from Figs. 7–9. This paper is just an initial report and a more complete analysis for different geographic regions and seasons, and the inclusion of additional CO_2 data sets for comparison, will be illuminating.

4. Conclusions

CO_2 measurements are very sparse in many parts of the globe such as the Southern Ocean (south of 30°S [30]) and in the upper troposphere and lower stratosphere. ACE-FTS observations help fill this gap and can contribute to carbon cycle science. ACE CO_2 VMRs are biased low at 5.5 and 6.5 km, but trend values are generally in good agreement with CT2017 and Toon's empirical CO_2 model. Based on model comparisons, the ACE CO_2 VMR values seem to decline too rapidly with altitude from 9.5 to 13.5 km. The ACE CO_2 product looks promising in this initial evaluation, but a full global bias assessment is necessary before ACE data can be assimilated into models such as CarbonTracker. ACE data is freely available upon request at http://www.ace.uwaterloo.ca/.

Acknowledgments

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Supplementary material

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jqsrt.2019.06.007.

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