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Vertically constrained CO_2 retrievals from TCCON measurements

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ABSTRACT

Partial column-averaged carbon dioxide (CO_2) mixing ratio in three tropospheric layers has been retrieved from Total Carbon Column Observing Network (TCCON) spectra in the 1.6 μm CO_2 absorption band. Information analysis suggests that a measurement with ~60 absorption lines provides three or more pieces of independent information, depending on the signal-to-noise ratio and solar zenith angle. This has been confirmed by retrievals based on synthetic data. Realistic retrievals for both total and partial column-averaged CO_2 over Park Falls, Wisconsin on July 12, 15, and August 14, 2004, agree with aircraft measurements. Furthermore, the retrieved total column averages are always underestimated by less than 1%. The results above provide a basis for CO_2 profile retrievals using ground-based observations in the near-infrared region.

1. Introduction

Remote sensing observations improve our understanding of the spatial and temporal distributions of carbon dioxide (CO_2) in the atmosphere. The Total Carbon Column Observing Network (TCCON) is a network of ground-based Fourier transform spectrometers (FTS). An automated solar observatory measures high-quality incoming solar absorption spectra in the near-infrared region ($4000-9000\ cm^{-1}$) (www.tccon.caltech.edu, [1,2]). Each TCCON instrument has a precise solar tracking system that allows the FTS to record direct sunlight. The absorption spectra are measured under clear skies and can be corrected by the recorded DC-signal for partly cloudy skies [3]. There are 20 sites located worldwide, including both operational and future sites. Although unevenly distributed over the world, the TCCON has good latitudinal coverage and the ensemble of sites retrieve the long-term column-averaged abundance of greenhouse gases, such as carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), and other trace gases (*e.g.* CO) with high accuracy and high precision [2,4–6].

The difference between column-averaged CO_2 (X_{CO_2}) and surface CO_2 can vary from 2 to 10 ppm or even larger depending on the location and the time of the year [7,8]. Higher surface concentrations usually occur at nighttime or in winter due to CO_2 build-up in a shallow planetary boundary layer (PBL), while surface uptake due to plant growth occurs during daytime or in summer. On both diurnal and seasonal time scales, the variations in X_{CO_2} are smaller than surface CO_2 because they are remotely forced by local fluxes through advection. Compared to surface values, the seasonal variation of X_{CO_2} generally has a time lag in phase with less variability due to the time delay caused by vertical mixing. The variations in X_{CO_2} are only partly driven by the local flux. Synoptic-scale activity has a large impact on the variations in X_{CO_2} due to larger-scale eddy fluxes and the meridional gradient in X_{CO_2}. The simulations by Keppel-Aleks et al. [9] illustrate that the sources of X_{CO_2} variations are related to the north-south gradients of X_{CO_2} and the flux on continental scales [9]. In contrast, the variations in PBL CO_2 are directly influenced by local flux [10]. They show that the PBL CO_2 variability is explained by regional surface fluxes related to land

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cover and mesoscale circulation across the PBL. In another study, Stephens et al. [11] concludes that most of current models overpredict the annual-mean midday vertical gradients and consequently lead to an overestimated carbon uptake in the northern lands and an underestimated carbon uptake over tropical forests [11]. Isolating the CO_2 dry air mole fraction in the PBL provides higher sensitivity to the surface-atmosphere fluxes since this is the portion of the atmosphere which is most strongly influenced by surface sources and sinks, and this is the layer which will have the largest changes in CO_2 concentrations from surface effects. The surface flux has a direct influence on the diurnal variations of the PBL CO_2. The mesoscale transport would also cause a horizontal difference in the PBL CO_2 concentration. Therefore, the estimates of the PBL CO_2 could provide regional scale constraints on the surface flux [10]. Additionally, multiple pieces of vertical CO_2 concentration information provide important constraints on atmospheric transport uncertainties when inverting these data for flux estimates.

In this paper, we show that high-resolution spectra of atmospheric absorption can provide information about the vertical distribution of CO_2 in addition to the total column abundance. Three scaling factors defined in three bulk atmospheric layers are retrieved to estimate the CO_2 vertical distribution. Other than the accurately retrieved total column abundance, the vertical variation is represented by the partial column averages in the three bulk atmospheric layers.

Major sources of uncertainty in the TCCON retrievals include those from spectroscopic data, measurement noise, instrument line shape function (ILS), temperature, surface pressure and zero-level offset. Significant effort has been undertaken to reduce the instrumental uncertainties of a TCCON experiment [3,12–15]. An overview of these uncertainties for TCCON measurements is discussed in [2,5]. The column measurements are calibrated and their precision is quantified using *in situ* aircraft profiles [5,16].

This study demonstrates the feasibility of retrieving multiple pieces of CO_2 vertical profile information from the high SNR, high spectral resolution atmospheric spectra obtained from TCCON. To ensure an accurate evaluation of the performance of our profile retrieval algorithm, we have limited our analysis to TCCON spectra acquired from the Park Falls, Wisconsin site, which coincides with *in situ* vertical profiles measured from aircraft. This comparison of ground-based and *in situ* data provides critical validation standards for retrievals of CO_2 profile and total column CO_2. This also enables an accurate error assessment. Our study provides a baseline to guide the next generation of remote sensing measurements for profiling the atmospheric CO_2. A more comprehensive evaluation of vertical profile retrievals from multiple TCCON sites for observing dates spanning nearly a decade and sampling a wider range of atmospheric conditions, seasons, and solar zenith angles is under way.

In the followings TCCON and aircraft data, information analysis, and the setup of the vertically constrained retrieval are described in the next section. An illustration of the profile retrievals using both synthetic and real spectra is discussed in Section 3, in which we use three of aircraft profiles, measured at Park Falls [4] on July 12, July 15, and August 14 in 2004, to evaluate the partial column retrievals from coincident TCCON spectra. The conclusions and discussions follow in Section 4.

2. Data and methodology

2.1. TCCON data

The TCCON X_{CO_2} measurements are precise to better than 0.25% [2,5]. With this precision, the monthly averaged column-integrated data are sufficient to reduce the uncertainties in the global surface carbon sources and sinks [17]. The absolute accuracy of the uncalibrated X_{CO_2} measurements from TCCON is $\sim$1% [5]. These measurements have been calibrated to 0.25% accuracy using aircraft profile data that are themselves calibrated to the World Metrological Organization (WMO) scale over nine TCCON sites (Park Falls, Lamont, Darwin, Lauder, Tsukuba, Karlsruhe, Bremen, Bialystok, Orleans) [5,18]. Consequently, they can be used in combination with *in situ* measurements to provide constraints on continental-scale flux estimates [9,19–22].

The TCCON X_{CO_2} measurements are an important validation source for satellite observations, including those from the Orbiting Carbon Observatory (OCO-2) [23], SCanning Imaging Absorption SpectroMeter for Atmospheric CartograpHY (SCIAMACHY) [24], and Greenhouse Gases Observing Satellite (GOSAT) [25–27]. In contrast to these space-based instruments, which measure reflected sunlight in the near infrared region by looking down from space, the retrievals using TCCON spectra have minimal influences from aerosol, uncertainty in airmass, or variation in land surface properties [2] because the TCCON instruments measure direct incoming sunlight from the ground. Thus, TCCON data serve as a transfer standard between satellite observations and *in situ* networks [1,2,5,6,28]. Because of their high quality, TCCON spectra are some of the best data to attempt the challenging profile retrieval.

2.2. Aircraft in situ profiles

The aircraft *in situ* measurements of CO_2 profiles have higher precision ($\sim$0.2 ppm) and higher accuracy ($\sim$0.2 ppm) [5] than the TCCON and spacecraft observations. We consider these measurements to be the best observations of the true state of the atmospheric CO_2 profile. In this study, the remote sensing measurements of CO_2 over Park Falls, Wisconsin on July 12, 15 and August 14, 2004 [4] are compared with the coincident *in situ* measurements during the Intercontinental Chemical Transport Experiment—North America campaign (INTEX—NA) [29]. Highly precise ($\pm$0.25 ppm) CO_2 profiles were obtained from 0.2 to 11.5 km in about a 20 km radius. Due to the altitude floor and ceiling limitations of the aircraft measurements, additional information for surface and stratospheric values of CO_2 are required. The lowest measured value is at $\sim$200 m above the surface, and it is assumed to be the surface value. The profile above the aircraft ceiling