

Carbon monoxide distribution from the ACE-FTS solar occultation measurements

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[1] This paper presents a comprehensive analysis of the CO observations acquired during the first eight months (January to September 2004) of the ACE mission. We show that the ACE high-resolution Fourier transform spectrometer (ACE-FTS), which operates in the solar occultation geometry and covers a wide spectral interval in the infrared, provides useful measurements in both the CO 1-0 and 2-0 vibrational bands. Vertically-resolved CO concentration profiles are retrieved, extending from the mid-troposphere to the thermosphere (from about 5 to 110 km). We have analyzed the latitudinal variability of the measurements, from which various physical and chemical atmospheric processes are highlighted for further study.
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1. Introduction

[2] Carbon monoxide (CO) is a key atmospheric species. In the troposphere, where it has a lifetime of several weeks to a few months [Logan *et al.*, 1981], it is a good tracer of pollution, and its observation allows the characterization of both emission sources and atmospheric motion of pollution plumes. In the remote troposphere, far from emission sources, CO is produced from methane oxidation, which contributes a mixing ratio of about 50 ppbv to the global background. The main sink for CO is chemical destruction by reaction with the hydroxyl radical (OH). It can also be transported across the tropical tropopause. In the stratosphere, CO is produced by the oxidation of methane by reaction with O(¹D), Cl, and OH and is converted to CO₂ by reaction with OH. Above 50 km, in the mesosphere and thermosphere, photolysis of carbon dioxide (CO₂) is the main source of CO, which reaches a concentration of 150–300 ppbv at 50 km and 5–20 ppmv at 80 km. At these altitudes, CO can be used to study the meridional circulation

and, in particular, the important seasonal pole-to-pole gradient [Solomon *et al.*, 1985].

[3] Ground-based atmospheric infrared measurements [Barret *et al.*, 2003] can be used to study tropospheric columns while millimeter-wave spectroscopy [de Zafra and Muscari, 2004] allows the routine monitoring of the mesospheric CO abundance. Currently there are extensive CO observations from space from a number of satellite platforms, which are yielding a global view of the CO tropospheric distribution [e.g., Clerbaux *et al.*, 1999; Beer *et al.*, 2001; Aumann *et al.*, 2003; Deeter *et al.*, 2003; Buchwitz *et al.*, 2004]. These nadir-looking instruments offer global coverage but provide limited vertical information. ATMOS [Rinsland *et al.*, 2000] and ISAMS [Allen *et al.*, 1999] were the first spatial instruments to measure CO in middle and upper atmosphere using occultation and limb geometry. More recently, CO profiles were measured by MIPAS/ENVISAT [Funke *et al.*, 2004], MLS/AURA, and the sub-millimeter instrument on the ODIN satellite [Dupuy *et al.*, 2004].

[4] This paper presents the CO measurements obtained during the first eight months of the Canadian Atmospheric Chemistry Experiment (ACE) space mission. ACE, which is similar in some respects to the NASA ATMOS experiment that flew 4 times on the Space Shuttle between 1985 and 1994, was designed to measure a suite of species involved in the complex ozone-related chemistry occurring at polar latitudes and to study the evolution of ozone depletion since the Montréal protocol and its amendments have entered into force. In this paper, the emphasis is placed on illustrating the capabilities of the ACE instrument to provide information on CO, as a tracer of pollution and convection in the lower layers of the atmosphere, and as a tracer of dynamics at high latitudes in the stratosphere and mesosphere. The ACE CO measurements are briefly described in the next section. Detailed information content and error budget analyses are provided to characterize the sensitivity of the measurements to the CO profile. The latitudinal distributions of CO derived from the operationally retrieved level 2 data are then presented and compared to model climatologies.

2. ACE Measurements and CO Retrievals

[5] The ACE mission was launched by NASA on 12 August, 2003. The satellite has a 74° inclined circular orbit at an altitude of 650 km and has both global and high latitude coverage. The principal ACE instrument is a high resolution (± 25 cm Maximum Optical Path Difference) infrared Fourier transform spectrometer (ACE-FTS) operating from 750 to 4400 cm⁻¹ in solar occultation mode [Bernath *et al.*, 2005]. ACE can observe a maximum of

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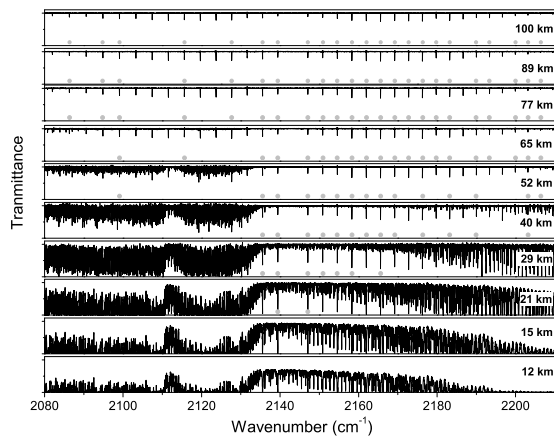


Figure 1. ACE-FTS occultation spectra for selected tangent altitudes, in the fundamental 1-0 absorption band. The transmittance scale is between 0 and 1. The microwindows used in the retrieval, varying with altitude, are identified by grey dots. Below 25 km, most of the information is retrieved from the 2-0 overtone band (not shown) as saturation and interferences due to other trace gases absorption increase when the instrument is sounding deeper in the atmosphere.

15 sunrises and 15 sunsets per day and the ACE-FTS measures infrared absorption spectra. These spectra are converted to atmospheric transmittance by division with an exo-atmospheric solar spectrum. The transmittance spectra contain information on the different atmospheric layers crossed by the line of sight, and their analysis provides vertical profiles of temperature and atmospheric constituents. Validation activities are ongoing. Preliminary results for P, T and several trace gases vertical profiles are found to be in good agreement with ground-based measurements and other satellites observations [Walker et al., 2005; McHugh et al., 2005; Jin et al., 2005].

[6] The CO molecule absorbs infrared radiation in the fundamental 1-0 and overtone 2-0 rotation-vibration bands, located near 4.7 and 2.3 μm , respectively. The two bands are covered by the ACE-FTS. Because a large dynamic range of optical thicknesses is encountered during a sequence of measurements, the CO retrieval can best be performed using transmittance information from both absorption bands. Figure 1 illustrates that the CO 1-0 band is clearly visible at high altitudes, but that the lines saturate at lower tangent heights. This saturation prevents an accurate tropospheric CO retrieval using this spectral region only. In the operational treatment of the ACE occultations, which uses a global-fit method in a general non-linear least squares minimization scheme (C. B. Boone et al., Retrievals for the Atmospheric Chemistry Experiment Fourier transform spectrometer, submitted to *Applied Optics*, 2005, hereinafter referred to as Boone et al., submitted manuscript, 2005), a varying set of microwindows in the fundamental band and in the overtone band are considered, for different tangent altitudes, to retrieve CO. The use of the intense 1-0 band provides information on the upper parts of the atmosphere where the absorption signal is weak, whereas the 2-0 lines become useful below around 25 km when the signal in the fundamental band saturates. The simultaneous use of the 1-0 and 2-0 bands offers the possibility of sounding CO

from about 5 km in the troposphere in clear-sky conditions to about 110 km in the thermosphere.

[7] The operational treatment of the ACE data does presently not provide the required information to perform a detailed a posteriori characterization of the retrieved CO profile. In the frame of this study, this has been achieved by retrieving the CO profile for a typical occultation, using the same microwindows but a different algorithm, based on the Optimal Estimation Method (OEM) [Rodgers, 2000]. Particular care has been taken to approach the conditions of the operational retrievals: The a priori covariance matrix was set to correspond to 50% variability of the CO content at all tangent altitudes, without any correlations between the layers. The analysis of the resulting averaging kernels shows that the measurements are very sensitive to the tangent altitude and that the vertical resolution varies between 1.4 km in the mid-troposphere to 3.9 km in the mesosphere (Figure 2 (left)). The sensitivity is the highest in the middle stratosphere, decreases in the mesosphere, and becomes important again above 80 km.

[8] The OEM also provides an efficient tool to quantify the error sources on the retrieved CO profiles. The total error for the occultation given as example in Figure 2 is of the order of 1–3% (Figure 2 (right)), except at low altitudes, around 60 km, and above 100 km, where it reaches 10–30%. It is calculated that the total error is mainly due to the statistical measurement noise, with additional contribution due to the smoothing of the true profile by the averaging

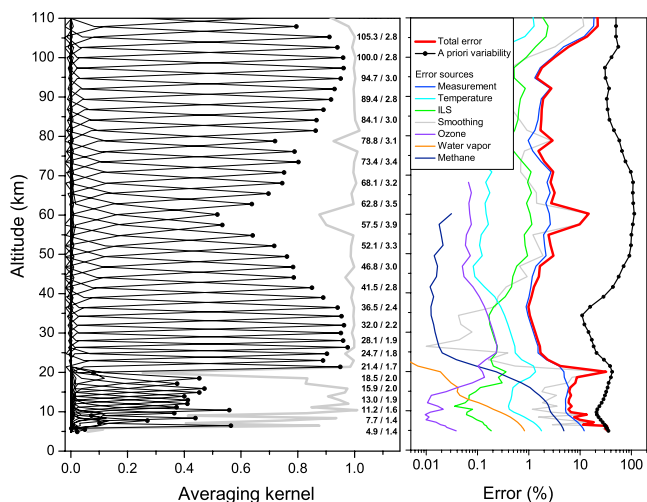


Figure 2. (left) Averaging kernels calculated for an occultation recorded over the Southern Hemisphere tropical region, on August 10, 2004. The peak of each averaging kernel gives the altitude of maximum sensitivity whereas its full width at half maximum can be interpreted as the vertical resolution of the retrieval; the corresponding values, in kilometers, are given for several averaging kernels on the right-hand side. Plain circles represent the nominal altitude of each kernel and the envelope (grey line) indicates the measurement response. (right) Error profiles for the same occultation. The total error and the individual contributions due to the measurement noise, to the smoothing and to uncertainties on the temperature, water vapor, methane and ozone vertical profiles, and on the instrumental lineshape, are plotted against the CO variability from model climatologies.

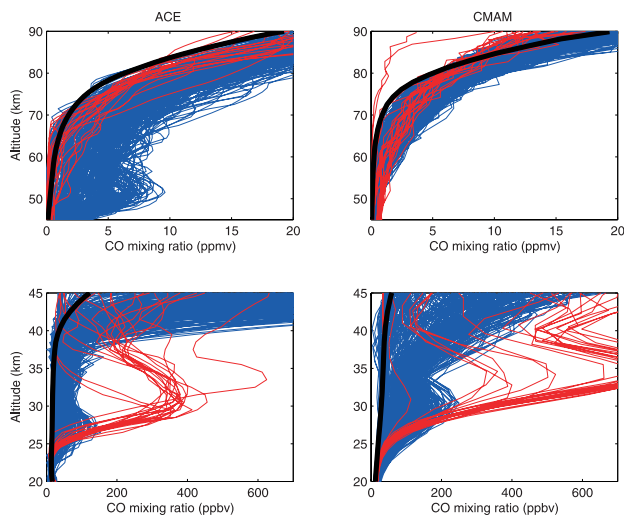


Figure 3. CO mixing ratio profiles (left) as measured by the ACE-FTS instrument and (right) as calculated by the CMAM model at the same location, in (bottom) the stratosphere and (top) the mesosphere. The thick black line is an average of all the profiles between 65°S and 65°N (700 occultations), corresponding to atmospheric conditions for which photodissociation of CO₂ regulates the CO content. The blue vertical profiles (312 occultations in January, February and March, poleward of 65°N), and the red vertical profiles (25 occultations in July and August, poleward of 65°S) illustrate that, at high latitude in winter, CO-rich air is transported downwards. Discrepancies are maximum between 30 and 50 km, where factors of 2 of disagreement are observed.

kernels matrix, as expected from the averaging kernels analysis. From Figure 2 (right), it can also be inferred that a little contribution is introduced by uncertainties on the retrieved temperature profiles and the profile of the main interfering species (H₂O, O₃, CH₄) in the troposphere, and by the Instrument Line Shape (ILS) function in the mesosphere and the thermosphere.

3. Vertical and Latitudinal Variability of CO Measurements

[9] In the following, we rely on the Level 2 (version 1) CO operationally retrieved data products (Boone et al., submitted manuscript, 2005) to study the atmospheric CO distributions. The preliminary validation of these data with measurements provided by the ODIN satellite report on an excellent agreement [Jin et al., 2005], thereby confirming the very high accuracy of the ACE measurements in the altitude range common to both instruments (20–100 km). In this section, we exploit all the available data (over 1000 profiles), from January to August 2004, to study the CO variability over the altitude, the latitude and the season.

[10] In order to show the large changes in mixing ratio due to transport processes occurring in the upper atmosphere and the usefulness of having regular spaceborne soundings to improve their understanding, the ACE vertical distributions are compared with the climatological profiles provided by the Canadian Middle Atmosphere Model (CMAM) [Beagley et al., 1997; de Grandpré et al., 2000]

at the same geographic location (see Figure 3). CMAM is a general circulation model with comprehensive chemistry of the middle atmosphere, that was run with a T32 resolution (about 500 km horizontally) and over 65 vertical levels. It can be seen that, for standard atmospheric conditions, when CH₄ oxidation and CO₂ photodissociation regulate the CO content, the model reproduces well the vertical distribution of the measurements (Figure 3, averaged black profile). On the contrary, when atmospheric dynamics is driving the CO concentration, at high latitudes in winter conditions, important discrepancies are observed (Figure 3, red and blue sets of profiles). In particular, it can be seen that between 25 and 55 km the amplitude and altitude of the downward transport of air occurring at high latitudes in winter differ from the CMAM prediction. The present observations of the CO profiles by ACE bring thus further evidence of the very disturbed character of the Arctic winter 2004, highlighted in several other publications [Manney et al., 2005; Nassar et al., 2005; Jin et al., 2005].

[11] Figure 4 depicts the latitudinal average (1° grid) of the available data, between 0 to 80 km, for the northern hemisphere winter period (January–March). Several features can be identified using these first ACE global measurements. As expected, in the mid-troposphere, one observes increasing CO concentrations at higher northern latitudes due to the presence of the main emission sources. The transition between concentrations characteristic of the upper troposphere (40–50 ppbv) and of the stratosphere (20 ppbv) is varying latitudinally in good agreement with the thermal tropopause. In the tropical tropopause layer, high CO levels are also measured as a result of convective transport from lower altitudes. In the upper stratosphere and the mesosphere, the strong gradient in CO concentration due to the meridional circulation, with upward transport in the summer polar regions, and downward transport in the winter high latitude regions of the northern hemisphere, is clearly observed.

4. Conclusions and Future Work

[12] In this paper we have highlighted the potential of the ACE-FTS instrument to measure CO profiles, from the mid-

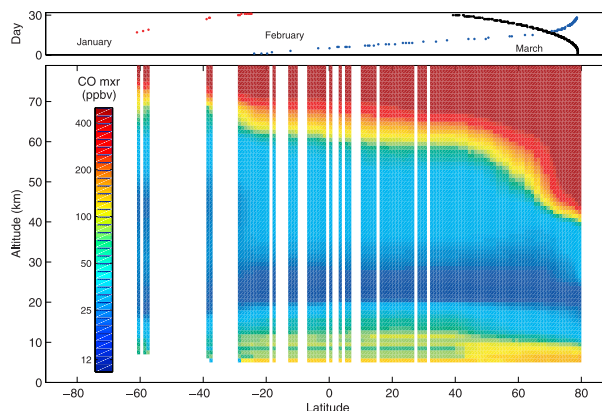


Figure 4. Latitudinal averages of CO mixing ratio profiles observed by ACE-FTS for the January to March 2004 period (corresponding to 508 occultations), plotted with a logarithmic colorscale.

troposphere to the thermosphere with a good accuracy. We have also demonstrated, by exploiting 8 months of available ACE-FTS measurements, that the study of the vertical and latitudinal CO distributions obtained by ACE provides valuable insights into the atmospheric dynamics. In the troposphere, the expected North-South hemispheric gradient has been identified and the effects of vertical transport due to convection in the tropical tropopause has been observed. In the strato-mesosphere, by comparing the observations with co-located model climatologies, we have shown that the ACE-FTS observation of CO as a tracer of the polar vortex dynamics is providing new information.

[13] These results open promising perspectives for further scientific studies on CO using the ACE-FTS satellite data. These should benefit from the increase of data downlink capabilities, which will enable more occultations to be measured. Improvements will also certainly be made with the version 2 of the operational ACE-FTS retrieval algorithm, which will include additional microwindows in the 2-0 band, in order to increase the accuracy in the troposphere. The capability for probing this part of the atmosphere is very valuable, as current space-borne instruments are limited to sounding either the mid-troposphere or the stratosphere and above. In conjunction with the other ACE data and with model studies including data assimilation, the measurements will give an integrated picture of polar chemistry and dynamics, and will help in understanding the evolution of ozone depletion.

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