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Co-located ACE-FTS and Odin/SMR stratospheric-mesospheric CO 2004 measurements and comparison with a GCM

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[1] This paper presents a comparison of co-located and near simultaneous CO measurements from January to May, 2004 and from the Arctic to southern polar regions using the ACE-FTS, in solar occultation mode, and the Odin/SMR, which measures atmospheric emission. We find that there is excellent agreement between the two instruments at the locations investigated over 4 orders of magnitude from the lower stratosphere to the lower thermosphere. There is also good agreement with the CMAM model simulation from 20 km to 90 km in sub-tropical and tropical latitudes but poorer agreement in the upper stratosphere and lower mesosphere in winter polar regions. For the Arctic in March 2004 this can be attributed, at least partly, to the unique dynamical processes in the stratosphere in the winter of 2003–2004. Clearly CO measurements from these instruments will provide a useful tool for testing model transport from the troposphere to the thermosphere.
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1. Introduction

[2] Carbon monoxide (CO) is an important atmospheric species with a chemical lifetime varying from several weeks to a few months, or even longer, and so is a useful tracer of atmospheric motions [Solomon et al., 1985; Allen et al., 1999]. In the stratosphere, CO is produced by the oxidation of methane (CH₄) while the reaction with OH is the main gas phase loss process. Above about 50 km, in the mesosphere and thermosphere, photolysis of CO₂ is the main source of CO.

[3] While there have been extensive measurements of tropospheric CO measurements in the last few years [e.g., Deeter et al., 2004], there have been fewer observations of

middle atmosphere CO. Mesospheric CO has been measured from the ground by millimeter-wave spectroscopy [e.g., Clancy et al., 1984; Forkman et al., 2003]. The ATMOS FTS on the Space Shuttle measured middle atmosphere CO profiles [Rinsland et al., 2000] using the solar occultation technique. The ISAMS (Improved Stratospheric and Mesospheric Sounder) instrument, which operated for about a year on the Upper Atmospheric Research Satellite (UARS) [Allen et al., 1999], used mid-infrared CO emission which required detailed non-LTE calculations to retrieve ground state CO densities. The SMR (submillimetre radiometer) instrument on the Odin satellite has also reported CO measurement in the middle atmosphere region [Dupuy et al., 2004].

[4] The Atmospheric Chemistry Experiment (ACE) is a Canadian satellite mission (SCISAT-1) for remote sensing of the Earth's atmosphere. The satellite was launched on 12 August, 2003 into a low Earth orbit (altitude 650 km, inclination 74°) with a primary mission to study the Arctic polar ozone loss in the winter and spring seasons. The primary instrument is a high spectral resolution (0.02 cm⁻¹) Fourier Transform Spectrometer (ACE-FTS) operating from 2.3 to 13.3 microns (750–4400 cm⁻¹) designed to measure a suite of species involved in the complex ozone-related chemistry occurring at polar latitudes [Bernath et al., 2005]. The primary working mode for ACE-FTS is solar occultation with a nominal vertical resolution of 3 km. Routine ACE science measurements began in February 2004.

[5] Validation of species profiles is an important and ongoing exercise for a satellite instrument. Although the occultation technique, such as used by the ACE-FTS and ATMOS instruments, is “self-calibrating”, it is nevertheless important to compare with other techniques. Additionally, because of the dependence of CO on transport properties of the atmosphere, it is useful to compare with models in order to evaluate their performance.

[6] In this paper we focus on a comparison of CO measurements with those from the Odin/SMR instrument using a set of co-located measurements at various locations and seasons.

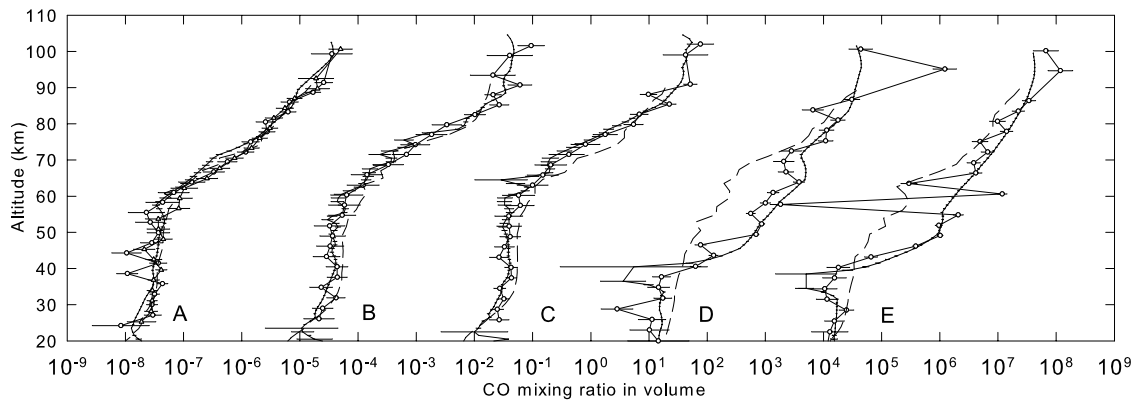


Figure 1. Plots of CO ACE-FTS (solid line) and SMR (solid lines with circles and triangles) observations for the 30 January, 2004, 7 and 8 April, 2004, and 16 and 17 May, 2004 with the coincidence as defined in the text. Also shown are CMAM calculations (dashed line) for the grid point nearest to ACE location for a single model year. Uncertainties provided for the ACE volume mixing ratio results are statistical errors from the fitting process ($1-\sigma$), and do not include systematic contributions. (A) ACE, (28.0°S, 329.0°E) 20:56 30 January, SMR, (30.3°S, 293.3°E) 10:24 30 January, triangles, SMR, (21.3°S, 295.4°E) 10:21 30 January, (B) ACE, (4.0°S, 42.3°E), 15:10 7 April, SMR, (0.6°N, 36.2°E) 15:52 7 April, (C) ACE, (9.8°S, 156.7°E) 07:29 8 April, SMR, (9.4°S, 157.1°E) 07:54 8 April, (D) ACE, (63.8°S, 172.4°E) 03:23 17 May, SMR, (60.1°S, 178.3°E) 10:20 17 May, (E) ACE, (64.1°S, 80.9°E) 09:28 16 May, SMR, (65.2°S, 63.3°E) 15:14 16 May, 2004. Profiles B, C, D, and E are scaled by 10^3 , 10^6 , 10^9 , and 10^{12} , respectively.

We note that we have compared individual profiles from both instruments and so, in some sense, this might be described as an acid test. This should serve as a cross evaluation of the measurement techniques used by each mission. We also compare the CO measurements with the CO field calculated by the Canadian Middle Atmosphere Model (CMAM).

2. Measurement Details

[7] The ACE-FTS measures CO by absorption of solar radiation in the (1-0) and (2-0) rotation-vibration bands located near 4.7 μm and 2.3 μm , respectively. The CO (1-0) band carries information on higher altitudes, while the weaker (2-0) band is used for lower altitudes. The retrieval process for ACE (C. D. 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) uses a generalized non-linear least squares minimization scheme while performing a global-fit to a set of micro-windows. For CO, thirty micro-windows in the fundamental band and 10 micro-windows in the overtone band are used simultaneously [Clerbaux et al., 2005]. Here we use the version 1 retrievals of Boone et al. (submitted manuscript, 2005) which provide volume mixing ratios with statistical uncertainties.

[8] The SMR on Odin satellite uses four tunable heterodyne receivers in the frequency range 486–580 GHz to perform limb measurements of thermal emission from trace constituents during day and night. CO is one of the target species of the Odin/SMR “odd hydrogen mode” together with H_2O , O_3 and HO_2 [Murtagh et al., 2002]. Odin has performed CO measurements since October 2003 on 2–3 observation days per month, time-shared with other aeronomy and astronomy observation modes. The target frequency band contains intense lines of CO at 576.268 GHz and O_3 at 576.515 GHz. The details of the CO inversion process have been presented by Dupuy et al. [2004]. The SMR CO retrievals were performed using

version 5.32 of the MOLIERE forward- and inversion model [Urban et al., 2004].

3. Results and Discussion

[9] Given the logistical constraints of the Odin operations, global vertically extensive CO measurements occur infrequently, whereas ACE CO measurements are continuous with nominally 15 occultations per hemisphere per day. However, due to downlink limitations not all of these are obtained. There are several coincident SMR and ACE-FTS observations during 2004: on 30 January at about 30°S, on 5 and 6 March at about 80°N, on 7 and 8 April in the tropics, and on 16 and 17 May at about 63°S. We note that while the SMR gives global coverage during a daily set of orbits, during the same period the ACE occultations occur around a latitude circle. The comparisons of the ACE-FTS and SMR CO data have thus been chosen on the basis of the following criteria for spatial and temporal coincidences. We consider it a spatial coincidence if the two observations are within about 4° of latitude and 10° of longitude, except for in January, when the longitudinal difference was relaxed to 40°. Time was not assumed to be a key factor but time differences are generally less than 7 hours and frequently much less.

[10] The occultation (A) in Figure 1 shows the results of a comparison of the ACE-FTS and SMR profiles for a single coincidence on the 30 January, 2004 at about 30°S. The measurements indicate volume mixing ratios of 10^{-5} to 10^{-4} in the mesosphere and lower thermosphere rapidly decreasing to about 3×10^{-8} at about 55 km. Below this altitude the mixing ratio is approximately constant and $\sim 3 \times 10^{-8}$. What is immediately evident is that although the CO mixing ratio spans over 4 orders of magnitude, there is overall excellent agreement between the profiles. The SMR profiles appear to be noisier than the ACE profiles as a result of measurement noise. The $1-\sigma$ error bars are indicated, and some stronger oscillations present in the version 223 data set are presumably caused by instabilities of the inversion

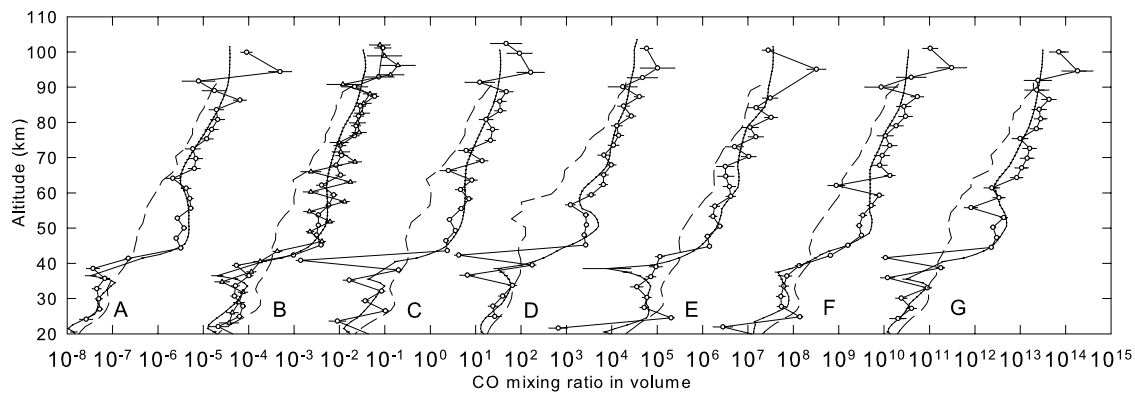


Figure 2. Results for 5 and 6 March, 2004 with details as for Figure 1 and capital letters referring to the occultations. (A) ACE, (79.5°N, 9.9°E) at 15:19 5 March, SMR, (77.4°N, 3.0°E) 15:34 5 March, (B) ACE, (79.5°N, 321.6°E) 18:34 5 March, SMR, (82.1°N, 319.8°E) 15:37 5 March; triangles, SMR, (77.2°N, 315.4°E) 18:47 5 March, (C) ACE, (79.5°N, 297.4°E) 20:12 5 March, SMR (79.1°N, 288.5°E) 14:03 5 March, (D) ACE, (79.4°N, 225.0°E) 01:05 6 March, SMR (79.5°N, 218.1°E) 18:52 5 March, (E) ACE, (79.3°N, 79.4°E) 10:52 6 March, SMR (79.6°N, 74.3°E) 04:31 6 March, (F) ACE, (79.2°N, 6.8°E) 15:45 6 March, SMR, (80.1°N, 5.2°E) 09:20 6 March, (G) ACE, (79.4°N, 249.0°E) 23:28 5 March, SMR, (76.2°N, 246.4°E) 23:36 5 March, 2004. Profiles B, C, D, E, F, and G are scaled by 10^3 , 10^6 , 10^9 , 10^{12} , 10^{15} , and 10^{18} , respectively.

process. We have also included a second SMR profile for 21°S which is just outside our coincidence limits: it is again quite similar. The steep increase in the mesosphere is due to the CO_2 photolysis source while the relatively constant mixing ratio in the stratosphere reflects the CH_4 oxidation source [e.g., Solomon *et al.*, 1985; Allen *et al.*, 1999]. These different photochemical sources appear to define a distinct transition in the profile which, however, is also modified by transport (see below).

[11] The occultations (B) and (C) in Figure 1 show the ACE-FTS and SMR observations for the tropics on the 7 and 8 April, 2004. Although there are more ACE-FTS observations only these two profiles that satisfy our coincidence constraints are presented. The profiles are similar to the sub-tropical profiles shown as occultation (A) except that the transition region appears to be somewhat higher at about 65 km. At about 80 km, CO tropical mixing ratios are similar to those in both the sub-tropics (A) and the Arctic (see Figure 2). Again the agreement between the two techniques is excellent above 25 km.

[12] The occultations (D) and (E) in Figure 1 show two coincidences for ~60°S on the 16 and 17 May. At these southern latitudes in late fall the transition region occurs at about 40 km while lower stratospheric mixing ratios are less than $\sim 2 \times 10^{-8}$.

[13] The seven panels in Figure 2 are for the vicinity of 80°N on the 5 and 6 March. The structure of the profiles while generally similar to those shown in Figure 1 also shows important differences. The mesospheric mixing ratio slope is less steep and there is a “ledge” at about 50 km with a rapid decrease to about 3×10^{-8} below.

[14] Dupuy *et al.* [2004] have compared the SMR CO retrievals for November 2001 with the ISAMS results obtained for the same month but 10 years earlier. They found that, in general, the SMR results were lower than the ISAMS results by a factor of 5 to 10 above 0.5 hPa and between 50°N and 80°N. Although there are limitations with this type of single profile comparison, the good agreement between ACE-FTS and the SMR strongly sug-

gests that there may be a problem with the parameters used in the complex non-LTE calculations of the ISAMS inversion process. This clearly requires further work.

[15] We have also compared the measurements with the CMAM which is a spectral General Circulation Model (GCM). The version that we have used has a top at 6×10^{-4} hPa (roughly 95 km geometric altitude) [Beagley *et al.*, 1997] and incorporates radiation, gravity wave drag, as well as the standard processes in a tropospheric GCM. CMAM also includes a comprehensive representation of middle atmosphere chemistry [de Grandpré *et al.*, 2000]. The horizontal resolution is T32 and there are 65 vertical levels. We note that the CMAM uses a fixed mixing ratio for CO_2 throughout the atmosphere and this could yield too high CO mixing ratios at the top of the model. However, we consider that it is quite likely that this condition, in some measure, will compensate for the downward flux of CO at the model upper boundary which has not been included.

[16] In the tropics and sub-tropics (Figure 1) we find very good agreement between the model and measurements. Generally the simulation is in the data range although the model results are slightly larger in the sub-tropics. Since CO_2 and CH_4 are the major CO sources above and below 50 km, respectively, this shows that CMAM has a good ability of simulating the oxidation of CH_4 and dissociation of CO_2 at low latitudes. For the late fall at high southern latitudes there is good agreement between model and the observations above about 75 km, while the model simulation is smaller than the measurements between 75 and 40 km, which suggests that there may be a problem with the polar descent of air in CMAM. Clearly this problem will require further examination.

[17] In the lower Arctic stratosphere, there is generally good agreement between the model and the measurements. In the upper Arctic mesosphere above about 70 km the difference between the model and the measurements is within a factor of two. Below ~60 km the discrepancy increases as the CMAM does not exhibit the ledge observed by both the ACE-FTS and the SMR instruments around 50 km: the observa-

tions are a factor of 5–10 greater than the CMAM simulation in this region. This is most likely due to a significant transport difference between the CMAM simulation and the real atmosphere. The arbitrary CMAM year used for the comparison does not have the features of the abnormal winter of 2003–2004 [Manney *et al.*, 2005]. We note that the ACE-FTS also observed dramatically enhanced values of NO in the lower mesosphere in February and early March 2004. This enhanced NO appears to result from the transport of NO produced by ionization in the lower thermosphere and upper mesosphere associated with solar proton events in October and November 2003 and the ensuing auroral activity associated with a disturbed magnetosphere [Rinsland *et al.*, 2005; Semeniuk *et al.*, 2005]. The longevity of the NO anomaly during its descent points to unusually persistent confinement of the NO to the polar night. This confinement was facilitated during January and February by the presence of a strong mesospheric polar vortex and this also resulted in downward transport of air with high mixing ratios of CO from the upper mesosphere and lower thermosphere.

[18] The strong mesospheric polar vortex was linked to the major stratospheric warming which developed by the middle of January 2004 and lasted for about a month. The disruption of the stratospheric vortex by planetary waves during the warming involves large scale mixing of polar air with air from lower latitudes [Allen *et al.*, 1999]. As a result, the high polar values of CO was diluted in the stratosphere. This sort of mixing did not occur in the mesosphere during this period, so a step-like feature in the vertical CO distribution was formed around the Arctic polar stratopause. The descent rate of CO in the upper stratosphere is too slow for the step-like feature to have been removed by early March. Analysis of the CMAM output (not shown) shows that there was much less polar confinement in the mesosphere compared to the observed atmosphere during 2004. In addition, the breakdown of vortex confinement was not reproduced in the stratosphere. So it should not be expected that the step-like transition in the late winter polar CO would develop during this CMAM winter or most winters in the real atmosphere.

[19] In summary, there appears to be excellent agreement over a large vertical range between the CO measurements from two instruments which use quite different techniques, one in absorption, the other in emission, which suggests that the ACE-FTS can offer reliable and high vertical resolution CO measurements in the middle and upper atmosphere along with the SMR instrument. Comparison of the observations with the CMAM shows good agreement at low latitudes, while poor agreement in the polar winter stratosphere suggests that there may be a problem with the polar descent rate in the CMAM. The large augmentation of CO at about 50 km in the measurements is due to the special dynamical processes that occurred in the winter of 2003–2004. Further analysis is required to elucidate model-measurement differences in the stratosphere. The use of CO observations from the ACE-FTS and SMR instruments is a very powerful tool to investigate transport in the middle atmosphere and should be exploited further.

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