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## Strato-mesospheric measurements of carbon monoxide with the Odin Sub-Millimetre Radiometer: Retrieval and first results

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[1] The *Sub-Millimetre Radiometer* (SMR) aboard the Odin satellite has been measuring vertical profiles of atmospheric trace gases since August 2001. We present the inversion methodology developed for CO measurements and the first retrieval results. CO can be retrieved from a single scan measurement throughout the middle atmosphere, with a typical resolution of  $\sim 3$  km and a relative error of  $\sim 10\%$  to  $\sim 25\%$ . Retrieval results are evaluated through comparison with data from the *Whole Atmosphere Community Climate Model* (WACCM) and observations of the *Improved Stratospheric and Mesospheric Sounder* (ISAMS) on board the *Upper Atmospheric Research Satellite* (UARS). Considering the large natural variability of CO, the SMR retrievals give good confirmation of the WACCM results, with an overall agreement within a factor of 2. ISAMS abundances are higher than SMR mixing ratios by a factor of 5–10 above 0.5 hPa from  $\sim 80^\circ\text{S}$  to  $\sim 50^\circ\text{N}$ . *INDEX TERMS*: 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 0394 Atmospheric Composition and Structure: Instruments and techniques. **Citation**: Dupuy, É., et al. (2004), Strato-mesospheric measurements of carbon monoxide with the Odin Sub-Millimetre Radiometer: Retrieval and first results, *Geophys. Res. Lett.*, 31, L20101, doi:10.1029/2004GL020558.

### 1. Introduction

[2] The Odin satellite is a Swedish-led international project involving Sweden, France, Canada, and Finland. The *Sub-Millimetre Radiometer* (SMR) on board Odin uses four tuneable heterodyne receivers in the frequency range 486–580 GHz to perform limb measurements of thermal emission from trace constituents during day and night. Observation time is equally shared between astronomy and aeronomy [e.g., Frisk et al., 2003]. The main targets of the aeronomy mission are trace gases relevant to middle atmosphere chemistry and dynamics such as O<sub>3</sub>, H<sub>2</sub>O and their isotopes, species related to ozone chemistry such as

ClO and HNO<sub>3</sub>, as well as long-lived tracers for transport processes, such as N<sub>2</sub>O and CO [Murtagh et al., 2002]. SMR measures the  $J = 5 \rightarrow 4$  rotational transition of CO at 576.268 GHz. The retrieval methodology and the first results are presented hereafter.

### 2. Carbon Monoxide Measurements

[3] Carbon monoxide, with respect to its role as a tracer for global scale transport [Solomon et al., 1985], is one of the target species of the Odin/SMR “odd hydrogen mode”, together with H<sub>2</sub>O, O<sub>3</sub> and HO<sub>2</sub>. In this mode, trace constituents are measured using two scanning modes. A mesospheric scan (55 to 110 km) is used to study H<sub>2</sub>O, CO and O<sub>3</sub> in the mesosphere. A strato-mesospheric scan (7 to 110 km) serves to investigate the coupling of the atmospheric layers [Murtagh et al., 2002].

[4] Priority for the 2001–2002 measurements was put on the “stratospheric mode” and the “water isotope mode” [e.g., Lautié, 2003; Urban et al., 2004a]. Sporadic CO observations were also performed during this period. We focus here on the strato-mesospheric CO observations on 7 August and 18 November 2001, which provide full altitude coverage. The target frequency band contains intense lines of CO at 576.268 GHz and O<sub>3</sub> at 576.515 GHz. The CO measurements use the full auto-correlator bandwidth of 800 MHz, with an effective resolution of 2 MHz [Olberg et al., 2003]. The altitude step between consecutive spectra is  $\sim 3$  km on 7 August. For 18 November 2001, it is  $\sim 1.5$  km below 60 km and  $\sim 5.5$  km above. The corresponding integration time for a spectrum is 2.6 s on 7 August, and  $\sim 1$  s below 60 km and  $\sim 4$  s above on 18 November.

### 3. Data Analysis Scheme

#### 3.1. Frequency Correction

[5] Due to a *Phase-Lock Loop* (PLL) failure, the local oscillator frequency of the 572 GHz radiometer is noticeably offset from the commanded frequency, thus inducing a frequency shift of the whole spectral band. This requires systematic correction before the retrieval process. Accurate frequency calibration is done by comparing the observed and theoretical center frequencies of the CO line. Results for 7 August show a positive frequency shift. The orbital mean value of the frequency shift is almost stable throughout the day (+149 to +153 MHz), and the variations during an orbit are quasi-sinusoidal, with a maximum deviation from the orbit mean of  $\pm 3$  MHz. On the other hand, the 18 November spectra exhibit a negative shift. The total variation over an orbit is maximal (+30 MHz) for the first orbit and smallest

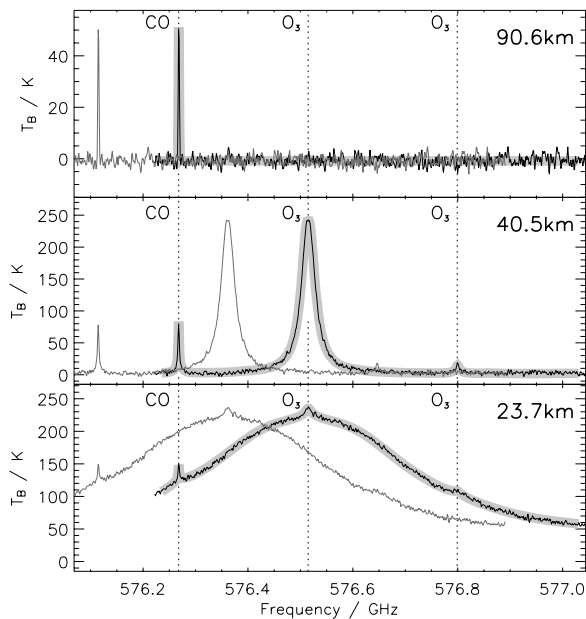
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**Figure 1.** Spectra obtained during a strato-mesospheric limb scan on 7 August 2001 ( $3^{\circ}\text{N}/120^{\circ}\text{W}$ ). Thin grey lines: observed spectra at 23.7, 40.5 and 90.6 km. The frequency corrected spectra (thin black lines) are superimposed with the reproductions calculated by the MOLIÈRE-5 code (thick grey spectra). Theoretical center frequencies are indicated by the dotted lines.

(+2 MHz) for the last one, with a single scan value decreasing from  $-105$  to  $-12$  MHz during the day. Finally, shift values for individual spectra of a single scan are separated by less than the spectrometer channel width (1 MHz), and the difference between the mean shift of consecutive scans is also lower than 1 MHz. The scan mean values of the shift were therefore used for correction of the calibrated spectra.

[6] Figure 1 shows spectra at 23.7 km, 40.5 km and 90.6 km, extracted from a limb scan near the Equator on 7 August 2001. The correction quality is illustrated by comparing the corrected spectrum (thin black) with the spectrum simulated by the forward model (thick grey), fully superimposed.

### 3.2. Retrieval Methodology

[7] A dedicated scheme has been developed and implemented in the level 2 processing chain CTSO (*Chaîne de Traitement Scientifique Odin*), version 223, employing the MOLIÈRE-5 (*Microwave Observation Line Estimation and REtrieval, version 5*) forward- and inversion model [Urban *et al.*, 2004b]. The non-linear retrieval scheme uses the *Optimal Estimation Method* (OEM) [Rodgers, 2000]. Volume mixing ratio (VMR) profiles of CO and  $\text{O}_3$  are retrieved simultaneously, along with the atmospheric temperature profile and the vertical profile of the continuum absorption. The inversion grid points of CTSO-v223 correspond to the tangent heights of the observations, to take into account variations of the resolution and tangent altitudes of Odin measurements. The a priori information for the retrieved species is obtained from the SMR climatology, based on monthly means of WACCM data in the

case of CO. The a priori pressure and temperature fields are taken from the *European Center for Medium-range Weather Forecasts* (ECMWF) below 45 km and from a model above. In addition, some instrumental parameters are retrieved. A pointing bias (of a few hundred meters), caused by errors in the attitude parameters and uncertainties on the effective pressure at the tangent point, is retrieved for each scan. A constant offset in brightness temperatures is estimated for each tangent height to account for positive or negative offsets present in the calibrated spectra. The retrieval of a residual frequency shift for each altitude ensures that frequency correction is performed correctly. Lastly, the spectroscopic parameters are taken from the Verdandi database, merging the JPL catalog and the broadening parameters from the HITRAN database (P. Eriksson, The Verdandi database, technical report, 2002, available at <http://www.rss.chalmers.se/gem/Research/verdandi.html>).

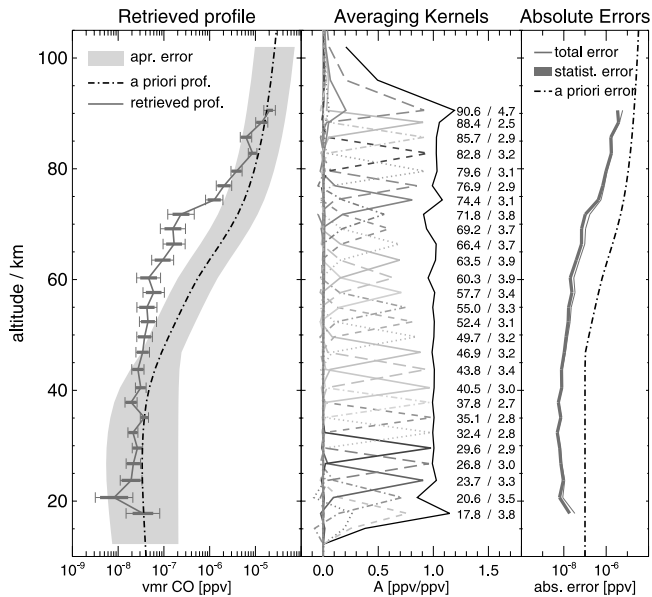
[8] Typical atmospheric values of the CO VMR range from a few ppbv to a few hundred ppmv. To improve numerical stability of the OEM scheme, the natural logarithm of the CO VMR is therefore retrieved. The absolute a priori error is set to 1.0 in logarithmic units, with a resulting minimum VMR error limited to 0.1 ppmv, applying in the stratosphere. For  $\text{O}_3$ , the a priori error is 75% of the ozone VMR, with a minimum value of 1 ppmv. A 5 K error is assumed for the temperature, with a minimum of 0 K. The  $\text{H}_2\text{O}$  continuum is retrieved logarithmically, with an a priori error set to 0.75 in logarithmic units and a resulting minimum VMR error of 1 ppmv. The noise in a spectrometer channel is given by the radiometer formula  $\Delta T_{rms} \approx T_{rec} / \sqrt{\Delta\nu \cdot \tau}$ . For a typical spectrum with a single-sideband receiver temperature  $T_{rec}$  of  $\sim 3500$  K, a resolution  $\Delta\nu$  of 2 MHz and a 2 s integration time  $\tau$ , we obtain a value of 1.8 K. A 3 K noise is added quadratically to the diagonal elements of the measurement covariance matrix, to account for variable instrumental effects which cannot be described by the forward model, such as baseline ripples.

[9] As an example, a retrieved vertical profile for the tropics on 7 August is shown on Figure 2. The single scan retrieval range, where the measurement response exceeds 80%, is  $\sim 18$ – $95$  km ( $20$ – $105$  km) on 7 August (18 November) 2001. It is limited by two factors: the low altitude signal is attenuated by absorption from the continuum and from the wings of strong out-of-band lines such as the  $\text{H}_2\text{O}$  line at 556.9 GHz; the upper boundary is constrained by the scanning limit. Vertical resolution on 7 August is  $\sim 3$  km between 25 and 40 km, and 3.5–4 km elsewhere. Typical resolution for 18 November is 1.9 km below 60 km and 5.5 km above. The differences are due to the variable integration times of the observations. The total retrieval error is dominated by the error due to statistical noise. An error value representative of the stratospheric part of the example profile is about 0.01 ppmv for corresponding retrieved VMR values of 0.02 to 0.06 ppmv.

## 4. First Results

### 4.1. Comparisons With WACCM and UARS/ISAMS

[10] SMR observations of CO on 18 November 2001 are compared with zonal averages of WACCM calculations for November and with the ISAMS measurements of November 1991 (Figure 3). WACCM is a General Circulation Model developed at the *National Center for*



**Figure 2.** Vertical profile retrieved from the spectra shown in Figure 1. Left: retrieved VMR profile (solid line) with total (thin) and statistical (thick) error bars. The a priori VMR and errors are also indicated. Center: averaging kernels. The full width at half maximum is given for each retrieval grid level. The envelope (black line) indicates the measurement response. Right: absolute retrieval error in ppv.

*Atmospheric Research* (NCAR). Its components were described in various papers [Roble and Ridley, 1994; Kiehl et al., 1998; Brasseur et al., 1998]. Forkman et al. [2003] found a good qualitative consistency between their ground-based CO measurements and the WACCM results. The WACCM original output is divided into 64 latitude bands and 66 pressure levels from  $\sim 1000$  to  $5 \times 10^{-6}$  hPa. ISAMS measured the infrared limb emission of the CO vibrational-rotational band at  $4.6 \mu\text{m}$ . The instrument was

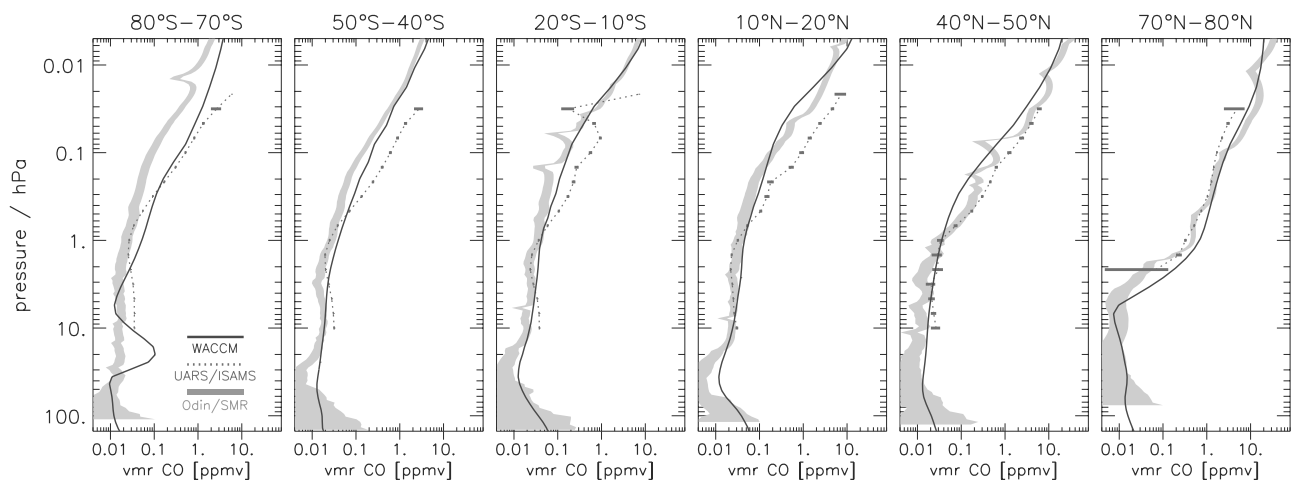
described in [Taylor et al., 1993], and the measurements in [López-Valverde et al., 1993]. We used version 10 of the level 3AT CO product. The ISAMS operational retrieval range for CO is 10 to 0.03 hPa.

[11] SMR zonal means on 18 November depart of one order of magnitude from the WACCM values at high latitudes of the summer hemisphere. A good agreement is found at other latitudes. A comparison for 7 August gives similar results (not shown). This is a satisfactory confirmation of the model by the SMR observations. The ISAMS and SMR CO mixing ratios are of the same order of magnitude for pressure values greater than 0.5 hPa (below  $\sim 55$  km). The agreement is best for northern mid- and polar latitudes. On the other hand, important discrepancies of a factor of 5 to 10 are found between SMR and ISAMS at pressure levels lower than 0.5 hPa. Furthermore, comparing the two SMR profiles of 18 November closest in longitude ( $\sim 10^\circ$ ) to the ground-based microwave measurements [see Forkman et al., 2003] taken on the same day at the Onsala Observatory ( $57^\circ\text{N}/12^\circ\text{E}$ ), we find VMR values from SMR greater than Onsala's by a factor of  $\sim 2$ . This is a quite good agreement, considering the geographical variability of the CO abundance since the edge of the polar vortex was close to Onsala at this time.

[12] The expected variation (up to 3 orders of magnitude) of the CO distribution, constrained by the interannual, seasonal and longitudinal variations of global scale transport, is enough in itself to account for the discrepancies of less than a factor of 10 found between the SMR, ISAMS and WACCM datasets.

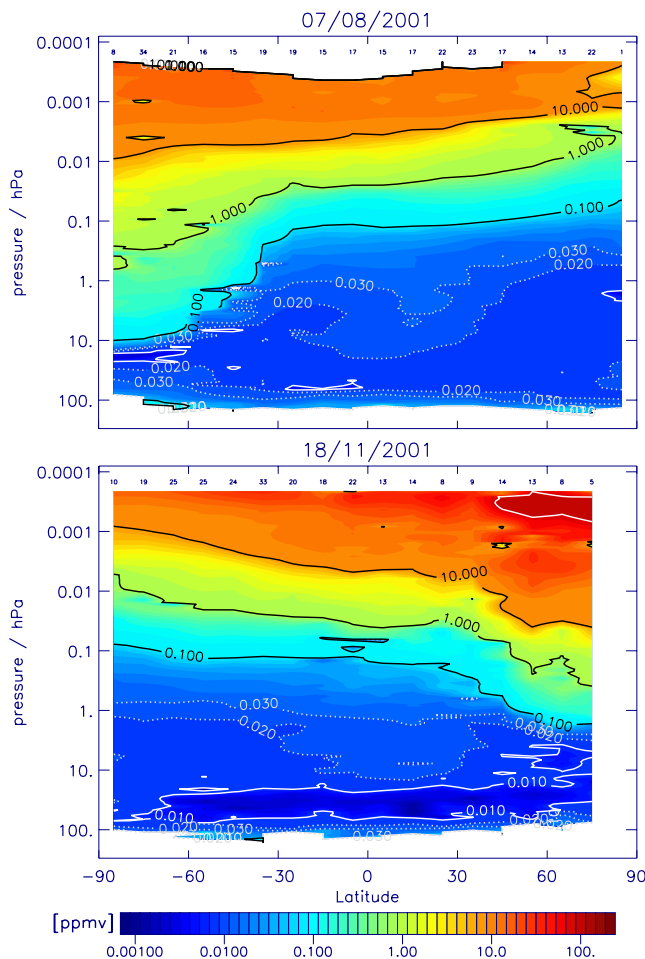
## 4.2. Global Distributions

[13] Daily zonal means for SMR measurements in 2001 (Figure 4) illustrate seasonal variations of the CO abundance. The major source of CO above the tropopause is  $\text{CO}_2$  photolysis in the thermosphere; a minor source is stratospheric  $\text{CH}_4$  oxidation. The main chemical sink for CO throughout the sunlit stratosphere is the recombination with OH [see, e.g., Allen et al., 1981].



**Figure 3.** Comparison of SMR CO zonal means for 18 November 2001 with: zonal means from ISAMS for November 1991, and zonal averages of WACCM data for the month of November. Shaded area: SMR  $\pm 3 \sigma$  error range. Dotted curve: ISAMS data with  $\pm 3 \sigma$  error bars. Solid line: CO climatology based on WACCM results. The vertical range is 200 to  $5 \times 10^{-3}$  hPa ( $\sim 15$  to  $\sim 85$  km). VMR values are plotted between 4 ppbv and 80 ppbv, in logarithmic scale.





**Figure 4.** Odin/SMR zonal mean CO for the strato-mesospheric observations of 7 August and 18 November 2001. Averaging is performed in  $10^\circ$  latitude bands. The vertical range is 200 to  $2 \times 10^{-4}$  hPa ( $\sim 15$  to  $\sim 105$  km). The VMR scale is logarithmic, from 1 ppbv to 400 ppmv.

[14] CO can be used as a tracer of global transport, as its photochemical lifetime (7 days at 50 km) is equivalent or longer than the vertical and horizontal advection timescales. Produced in the thermosphere, CO follows the global meridional circulation towards the winter hemisphere polar night region. Consecutive downwards motion induces a sharp gradient in CO concentrations down to the stratosphere [Solomon *et al.*, 1985], traced on Figure 4 by the 0.1, 1 and 10 ppmv contours. A value of 0.1 ppmv is found at a pressure level of 10 hPa ( $\sim 30$  km) on 7 August, comparable to ISAMS observations ( $\sim 35$  km) [Allen *et al.*, 1999]. Meanwhile, air masses with low CO content are uplifted through the summer stratosphere and mesosphere, explaining the lower VMR values found in the summer hemisphere.

[15] The efficiency of the CO+OH reaction and a low vertical diffusivity explain the global minimum observed around 50 hPa ( $\sim 22$  km) [e.g., Clancy *et al.*, 1984], highlighted by the 0.01 ppmv contour on the 18 November plot. The lowest value measured is  $\sim 4$  ppbv. The tropical stratospheric increase (0.02 and 0.03 ppmv dotted contours) is similar to the enrichment identified by Allen

*et al.* [1999] in the ISAMS data and attributed to methane oxidation.

[16] Finally, the seasonal reversal of global circulation explains the hemispheric anti-symmetry of the zonal means. Some differences remain between the dates, due to their respective position in the course of the seasons: 7 August is typical of the northern hemisphere mid-summer, whereas 18 November corresponds to the late fall/early winter period.

## 5. Perspectives

[17] Odin/SMR is the first instrument to provide global-scale daytime and nighttime microwave observations of CO, with good vertical resolution of 3–4 km in the whole middle atmosphere. Since a robust frequency correction and retrieval methodology is now available, measurements have been rescheduled to 2–3 days per month since October 2003, to allow for the construction of an accurate CO climatology based on SMR data. Even though the PLL seems to be operational since July 2002, the correction method presented here must still be used as a basic check to account for possible failures. Thorough validation of the SMR CO product, including a statistical study using the Onsala ground-based measurements, cross-comparison with the *Michelson Interferometer for Passive Atmospheric Sounding* (MIPAS) on board Envisat and the recently launched *Microwave Limb Sounder* (MLS) aboard Aura, will be addressed in a separate paper. Further work includes verification of model results concurrent with the SMR measurements, such as from 3-D *Chemical Transport Models*.

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