A joint retrieval of CO and vibrational temperature from MIPAS-ENVISAT

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Abstract

MIPAS is a limb viewing fourier transform spectrometer operating in the infrared. It scans the tangent altitude range 6-68 km at a vertical resolution of approximately 3km.

CO has various natural and anthropogenic sources including forest fires and industry. In the troposphere, CO is the main sink of the principal oxidising agent - the OH radical - and so reduces its capacity for the removal of other atmospheric pollutants. In the stratosphere, its intermediate lifetime makes it useful as a tracer for stratospheric motions and tropospheric-stratospheric exchange.

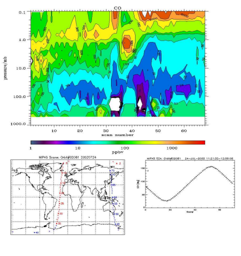
However, the retrieval of CO from a limb sounding IR instrument is complicated because of non-LTE effects. Ordinarily, the internal vibrational energy level populations are controlled by collisions between molecules and follow the translational (kinetic) energy distribution of the ambient gas (LTE). However, if collisions are infrequent, radiative processes can lead to a non-Boltzmann distribution of the internal energy level populations (non-LTE). CO is strongly affected by non-LTE in the IR down to around 40km and modelling these processes is difficult. These effects are especially problematic in the limb viewing geometry due to the long path length viewed through the upper atmosphere.

However, using a joint CO, vibrational temperature (T_v) retrieval it is possible to estimate the non-LTE effects without modelling the energy level populations. Instead, we retrieve a parameterisation of the non-LTE effects, T_v, directly from the emission spectra.

The non-LTE Problem

At high pressures collisions between CO and the ambient gas ensure the populations of the vibrational states of CO follow the kinetic energy distribution of the surrounding gas (Local Thermodynamic Equilibrium [LTE]). However, at low pressures, other processes dominate. During the day, CO is excited into higher energy states, mainly pumped by solar radiation. Few collisions mean that these states are maintained out of equilibrium with the surrounding gas (non-LTE). Collisions with other molecules in non-LTE are also important. At night this effect is reversed, as CO tends to radiate efficiently to space at high altitudes. These effects are important in the stratosphere and above.

The plot below shows what happens if we retrieve CO and do not account for non-LTE effects. The CO concentration itself has no diurnal variation. However, we see much higher values of CO VMR in the daytime scans at high altitudes.



A useful parameterisation of the non-LTE effects is the vibrational temperature, T_v. This describes the non-LTE population of a level m with energy E_m [5]:

$$T_{vm} = \frac{E_m}{k \ln\left(\frac{n_0 g_m}{n_m g_0}\right)} \qquad (1)$$

We need only consider the first excited vibrational state in the case of CO.

Retrieval Method

The forward model used is the RFM (Reference Forward Model) [2]. This model does not calculate the non-LTE populations. However, if the T_v profiles are known it is able to calculate the expected non-LTE radiances [3]. We can therefore treat T_v as another parameter to be retrieved from the emission spectra.

CO and T_v are retrieved simulataneously. The retrieval uses Levenberg-Marquardt Optimal Estimation (LMRTV). This uses prior knowledge about the expected atmospheric state and estimates the most probable state based on this *a priori* information in conjunction with the emission measurements. We use the retrieved kinetic temperature (T_k) profile as our *a priori* estimate of the T_v profile. Climatological values of CO are used as *a priori* for the VMR retrieval.

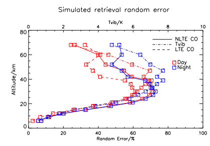
Retrieval Precision

Retrieval precision is calculated using an *a priori* uncertainty of 100% for CO and 10K for T_v as;

$$\mathbf{S_x} = (\mathbf{k^T S_y^{-1} k} + \mathbf{S_a^{-1}})^{-1} \qquad (2)$$

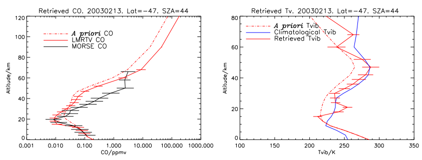
where $\mathbf{S_y}$ is the instrument noise covariance, $\mathbf{k}$ is the forward model jacobian and $\mathbf{S_a}$ is the *a priori* uncertainty covariance.

The plots below show the expected precision. We expect retrieval random errors for CO of better than 70% at most altitudes and better than 20% below around 18 km.

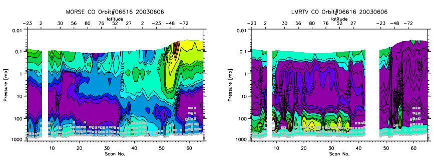


Retrievals from Real Data

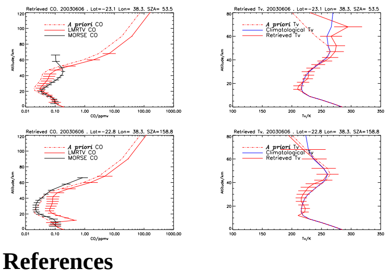
A comparison of the CO only LTE retrieval using MORSE (MIPAS Orbital Retrieval using Sequential Estimation) [1] and the new joint retrieval of CO and T_v using LMRTV from a daytime scan is shown below. The T_v climatological profile has been derived from a non-LTE populations model [4]. Using the joint retrieval the T_v profile is close to the climatological profile and the retrieved CO VMR is lower at high altitudes, as expected.



The following figures show orbit 06616 processed with MORSE and with LMRTV. The colour scale is the same in each plot. Daytime scans are 21–56. LMRTV removes the unrealistic enhancements.



The following figures show retrieved CO and T_v from coincident daytime and nighttime scans on 6th June 2003. We expect the daytime and nighttime retrieved CO to look similar. The MORSE profiles look very different between day and night. However, the LMRTV retrievals are more similar. Climatological daytime and nighttime T_v profiles are derived from a non-LTE population model [4]. The retrieved T_v profiles look roughly as expected.



References

[1] A. Dudhia. *MIPAS Orbital Retrieval using Sequential Estimation*. www.atm.ox.ac.uk/MORSE.

[2] A. Dudhia. *Reference Forward Model*. www.atm.ox.ac.uk/RFM.

[3] D. P. Edwards and M. López-Puertas. Non-Local Thermodynamic Equilibrium Studies of the 15-μm Bands of CO_2 for Atmospheric Remote Sensing. *Journal of Geophysical Research*, 98, 1993.

[4] M. López-Puertas, S. Gil-López, B. Funke, T von Clarmann, N. Glatthor, S. Kellmann, M. Milz, T. Steck, G. P. Stiller, A. Dudhia, V. Payne, V. Jay, J. Reburn, H. Bovensmann, and A. Bracher. Advanced MIPAS Level 2 Data Analysis (AMIL2DA). Technical report, Instituto de Astrofisica de Andalucia, 2003.

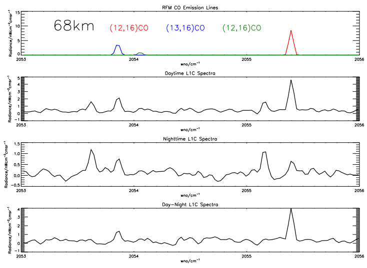
[5] M. López-Puertas and F. W. Taylor. *Non-LTE Radiative Transfer in the Atmosphere*. World Scientific, 2001.

Vibrational Temperature of CO Isotopologues

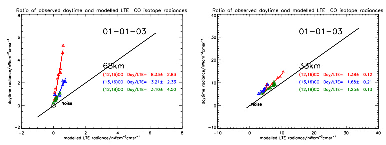
The non-LTE characteristics of the weak isotopic lines are not well understood but may affect the CO retrieval. CO isotopologues in the Earth's atmosphere in order of abundance are:

$$^{12}C^{16}O \quad ^{13}C^{16}O \quad ^{12}C^{18}O$$

By averaging 1050 spectra from a single day (01-01-2003), isotopic lines are become visible.



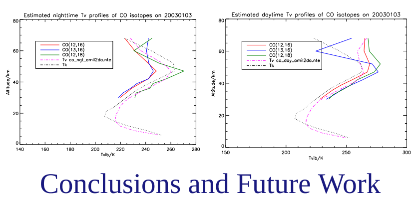
The plots below show the observed radiance plotted against the RFM modelled LTE radiance. At 68 km the observed radiance is enhanced with respect to its LTE value due to non-LTE effects. The non-LTE enhancement differs between isotopes. At 33 km the observations are closer to the modelled LTE radiance.



If we assume the atmosphere is optically thin then we can derive T_v profiles from;

$$\frac{R_{non-LTE}}{R_{LTE}} = \frac{\exp\left(\frac{h\nu}{kT_k}\right) - 1}{\exp\left(\frac{h\nu}{kT_v}\right) - 1} \qquad (3)$$

The figures below show the T_v profiles obtained. For the 1st and 3rd most abundant CO isotopologues, the daytime T_v derived using this method are quite close to the T_v climatology taken from a non-LTE population model [4]. However, the 2nd most abundant differs significantly at 60 km, which requires further investigation. The nighttime T_v are slightly different from the climatological T_v. These results suggest that T_v of the less abundant isotopologues may be slightly different from T_v of $^{12}C^{16}O$.



Conclusions and Future Work

If we do not attempt to account for non-LTE, we retrieve unrealistically high values of CO in the daytime at high altitudes. However, we can perform a joint CO, T_v retrieval to account for these non-LTE effects. This does not significantly degrade retrieval precision below around 21 km. The joint retrieval produces T_v that is reasonably consistent with expected values and the retrieval of CO appears to be improved. Investigations into the CO isotopologues suggest that they may have slightly different T_v and we have been able to make an estimate of their T_v profiles.