

Technical note: nonLTE model validation using MIPAS high altitude spectra

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July 28, 2008

Abstract

One of the parameters used to determine the best possible subset of spectra, or microwindows, to retrieve temperature and gas concentrations from MIPAS is the vibrational temperature that estimates the nonLTE effects acting upon the radiances. A technique that uses the daytime and nighttime MIPAS midlatitude middle atmosphere radiances to evaluate the quality of the vibrational temperatures used on the microwindow selection procedure is described. This technique is applied to the transitions of CO₂, O₃, H₂O, HNO₃, CH₄, N₂O, NO₂, NO and CO encountered in the MIPAS spectral range.

1 Introduction

The inversion of temperature, pressure and gas concentrations from infrared measurements usually relies among other things on the assumption that the emitting species are in local thermodynamic equilibrium (LTE) (Gille and House, 1971; Rodgers, 1976, 2000). This assumption implies that their populations are determined by the Boltzmann distribution at the local kinetic temperature and therefore, the source function of the emission is given by the Planck function at this temperature.

This assumption is no longer valid at high altitudes for most of the molecules. At these high levels, the populations of the molecules deviate from the Boltzmann's law mainly due to the slower collision rates, solar pumping, chemical reactions and the absorption of radiation from lower atmospheric layers. These processes affect the source function which is no longer simply the Planck function at the local kinetic temperature. Models have been developed to deal with the radiative transfer equation under nonLTE scenarios (Lopez-Puertas et al., 1986a,b; Wintersteiner et al., 1992). The nonLTE models of Lopez-Puertas provide the vibrational temperatures used to estimate the nonLTE effects in the radiances observed by MIPAS. These vibrational temperatures are used to characterise one of the assumed error sources in the process of selecting optimal subsets (known as microwindows) of the MIPAS spectra used to retrieve atmospheric parameters (Dudhia et al., 2002).

The purpose of this study is to use MIPAS middle atmosphere measurements to validate the modelling of the nonLTE emissions. As already noted by Lopez-Puertas et al. (2005) the MIPAS instru-

ment has several advantages for studying nonLTE radiances such as: (1) its wide spectral range which allows the identification of the emission from the same molecule in different spectral regions, (2) its spectral sampling (0.0625 cm^{-1} for the reduce resolution mode) which enables the identification of individual emission lines for different gases and furthermore for different vibrational transitions of a particular gas, and (3) its special viewing modes that cover the entire mesosphere and part of the thermosphere where nonLTE emissions become more significant (the middle atmosphere mode covering the atmosphere from 18 to 102km and the upper atmosphere mode covering the atmosphere from 42 to 172km).

In the following sections a technique to validate the diurnal nonLTE effects rather than the absolute nonLTE emission is introduced. This technique relies on the ratio of the daytime to nighttime radiance assuming small nighttime nonLTE effects. Even though the technique presented here can be applied to any molecule, for clarity it will be explained first for CO_2 and then it will be applied to other constituents.

2 Methodology

The MIPAS instrument measures the atmosphere around 10:30 local time on the descending and around 22:30 on the ascending part of its orbit. The difference between descending (day) and ascending (night) zonal mean spectra results from diurnal variations in gas concentrations, temperature and nonLTE effects. When observing the emission of a well mixed gas with no diurnal variation such as CO_2 , the ratio between the day and the night radiances will depend on the small diurnal variation of kinetic temperature and on the nonLTE effects (e.g. the upper energy level of the vibrational transition is pumped by the solar radiation during day but not during night). This measured ratio can be compared against the ratio predicted by nonLTE models. Comparing the ratios rather than absolute differences allows any variation between the atmospheric and model kinetic temperature and volume mixing ratio (VMR) to be eliminated hence leaving mainly the nonLTE effects.

The simulated data are created using Reference Forward Model (RFM) for CO_2 for the same tangent heights measured by MIPAS for day and night midlatitude climatological profiles. These profiles include the vibrational temperatures calculated the nonLTE model of Lopez-Puertas for these two conditions and use the same kinetic temperature as well as CO_2 concentration for both scenarios.

The analysis starts by sorting the MIPAS middle atmosphere apodized spectra for midlatitudes north or south (ie. 30°N to 60°N or 60°S to 30°S). This latitude range is used because it avoids the equatorial region where there is a significant diurnal tide, it avoids high latitude regions where the atmosphere is more variable and furthermore, because we only have both day and night vibrational temperatures for midlatitude conditions. These sorted apodized spectra are then separated into day time and night time data using the Solar Zenith Angle (SZA) information. Both data sets are then averaged height by height to produce representative day and night mean MIPAS radiances for midlatitude north and south conditions.

2.1 Spectral masks

As already mentioned, MIPAS' wide spectral range enables the identification of the emission from a particular constituent in different parts of the spectra. As shown in Figure 1, CO₂ emissions are significant in the spectral regions corresponding to the A, AB and D MIPAS bands. To demonstrate the nonLTE effects the simulated night nonLTE emission are overplotted on top of the simulated day nonLTE emissions.

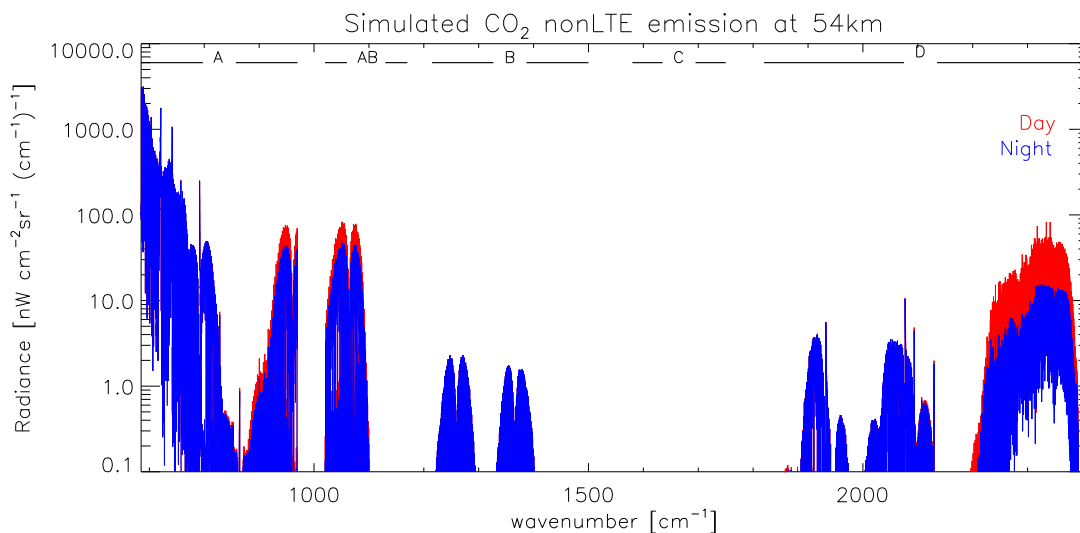


Figure 1: Simulated nonLTE CO₂ day and night emissions at 54km for the day atmosphere

Spectral masks for each tangent height are used to isolated emissions from CO₂ in the correspondent bands. These spectral masks are created as follows. In order to identify emission lines from CO₂ in the A band a spectrum that includes the emission of the strongest molecules for that particular spectral region (a “complete” spectrum) is simulated with the Reference Forward Model (RFM). Also, a CO₂-only spectrum is simulated and compared against the “complete” one. The emission lines that do not change by more than 10 % are assumed to be lines where CO₂ is the main contributor thus avoiding lines overlapped by other molecules. Figure 2 displays the “complete” simulated spectrum as well as the CO₂-only simulated spectrum for a subset spectral region of band A at 54km. The enhanced emission in the “complete” spectrum due to overlapping emission from other molecules rather than CO₂ is clearly visible around 708.70 cm⁻¹ wavenumbers. The light blue shaded regions correspond to the emission features identified as “pure” CO₂ lines with this method. Only lines that are above 10 % of the expected NESR (noise equivalent spectral radiance) for the measurements are used in order to avoid poor signal to noise (S/N) ratio. These masks are generated individually for all tangent heights between 18–90km.

Table 1 summarises the molecules used to generate the “complete” spectra for each band.

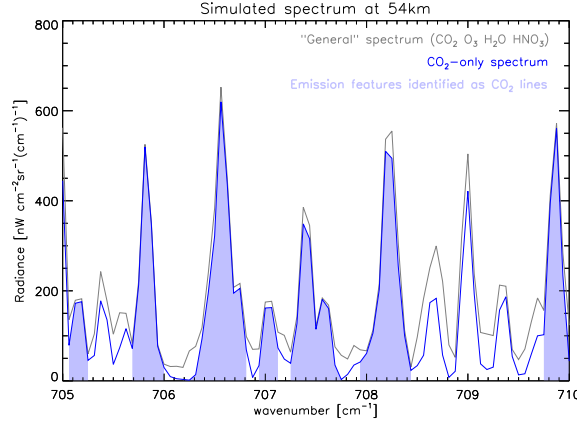


Figure 2: “Complete”, CO_2 -only spectrum and “pure” CO_2 emission features at 54km for a subset spectral region of MIPAS band A

2.2 Day/night ratio

These spectral masks were applied to the average midlatitude day and night spectra to discriminate the day and night CO_2 emission from the emission of the rest of the molecules observed by MIPAS and the ratio of these two “pure” CO_2 spectra was taken. This ratio is calculated using a weighted mean of the lines to minimise the influence of noise. The measurement ratio R_m is given by

$$R_m = \frac{\sum_i m_{di} s_{ni}}{\sum_i m_{ni} s_{ni}} \quad (2.1)$$

where m_{di} and m_{ni} correspond to the measured radiance of the i th “pure” CO_2 day and night midlatitude MIPAS emission feature and s_n correspond to the radiance of the i th CO_2 spectral line predicted by the nonLTE model using the night climatological profile.

This measurement ratio can be compared against the ratio predicted by the nonLTE model. This simulated ratio R_s is given by

$$R_s = \frac{\sum_i s_{di} s_{ni}}{\sum_i s_{ni}^2} \quad (2.2)$$

where s_{di} corresponds to i th CO_2 spectral line predicted by the nonLTE model when using the day climatological profile. In the case of CO_2 the only differences between the day and night atmosphere used to predict the simulated ratio R_s are the vibrational temperatures given by the nonLTE model. As an example, Figure 3 shows the day and night vibrational temperatures for some of the transitions

Table 1: MIPAS bands spectral ranges

Band	Molecules
A	CO_2 O_3 H_2O HNO_3
AB	CO_2 O_3 H_2O N_2O
B	CH_4 H_2O N_2O
C	NO_2 H_2O
D	CO_2 O_3 H_2O NO CO N_2O

of the main isotope in MIPAS band A. The transitions are written with the format CO₂ (L:U) where “L” and “U” correspond to the lower and upper HITRAN global quantum indices (i.e. vibrational indices) respectively.

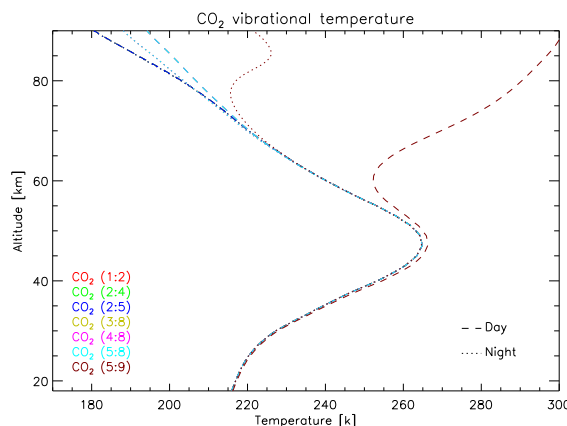


Figure 3: Day and Night vibrational temperatures for some CO₂ transitions in the MIPAS band A

3 CO₂ results

Figure 4 displays the result of the analysis for midlatitudes north and south for CO₂. A gray dashed line represents the position at which the day/night ratio is equal to one. When the day and night emission is in LTE or when the nonLTE emissions are not affected by solar illumination the expected result for the day/night ratio is one. Different days were analysed in order to allow for the effect of some SZA variation. Table 2 displays the mean SZA encountered in midlatitudes south and north for the day (d) and night (n) cases.

Table 2: Mean SZA variation

Day	Midlatitude North	Midlatitude South
20070422	d:39.8 / n:118.0	d:64.8 / n:141.6
20070607	d:32.0 / n:107.7	d:72.8 / n:150.3
20070824	d:42.5 / n:118.3	d:62.4 / n:141.6
20071013	d:57.7 / n:137.1	d:43.8 / n:124.2
20080215	d:63.8 / n:118.3	d:44.5 / n:117.5

The first result that can be noted from Figure 4 is that the measured ratio does not change much between midlatitudes north and south. It also can be seen that the CO₂ emission from band A and D is in general well modelled: the simulated ratio (the solid line) falls between the atmospheric variability encountered in the measured ratio (dashed lines with different symbols). A small deviation from the simulated ratio is visible around 62km for band A for all days at both midlatitudes. A similar deviation is encountered in the measured CO₂ ratio for band AB. Apart from this bump, this ratio differs significantly against the simulated ratio.

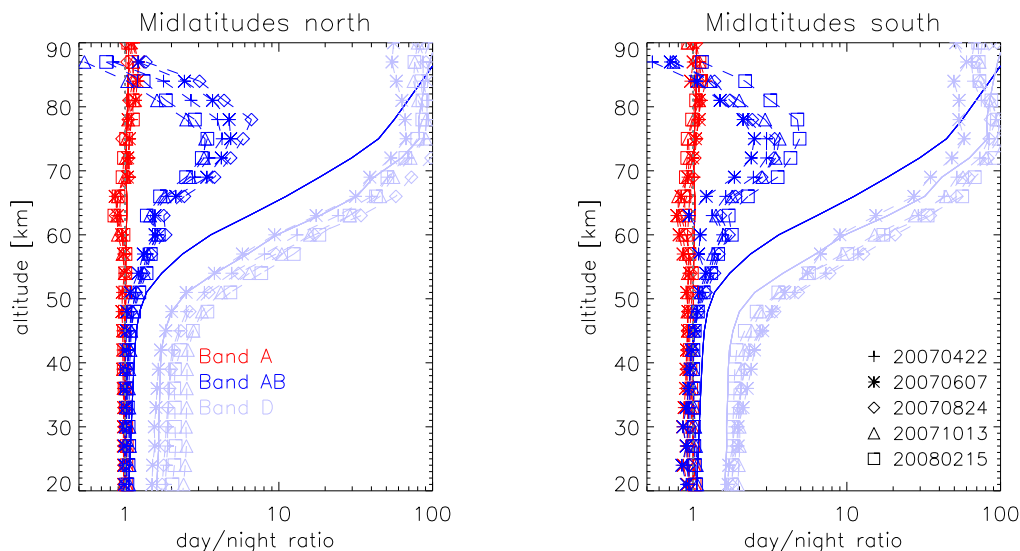


Figure 4: Simulated and measured day/night ratio for CO_2 for several days for midlatitude North and South scenarios. Dashed lines with different symbols represents the measured ratio for different days while the solid line corresponds to the simulated ratio.

4 TRANSITIONS

The analysis presented in the previous section can be extended to cope not only with the emission of CO_2 at different MIPAS bands, but also with the emission from different vibrational transitions for CO_2 . This implies identifying inside the already CO_2 masked spectra the lines which correspond to different transitions.

Once the strongest transitions were identified, transition spectral masks were created using the same procedure as the one described in subsection 2.1 in order to use them on top of the molecular spectral masks built previously.

Figure 5 displays the result of this extended analysis for midlatitudes south. The top panel shows the individual transitions identified for CO_2 in MIPAS band A, AB and D. The bottom panel shows the measured day/night ratio for the same days as in Figure 4.

Practically all the transitions in Figure 5 are in general well modelled. The exceptions are the transitions $\text{CO}_2(5:9)$ in band A and the transition $\text{CO}_2(3:9)$ in band AB. Both transitions have upper level “9” which suggests that discrepancy is a problem with the nonLTE model because the population of the upper level determines the emission from the transition. These transitions show a significant difference in magnitude below around 70km and in both magnitude and trend above 70km.

The same results are found when analysing the average midlatitudes north radiances.

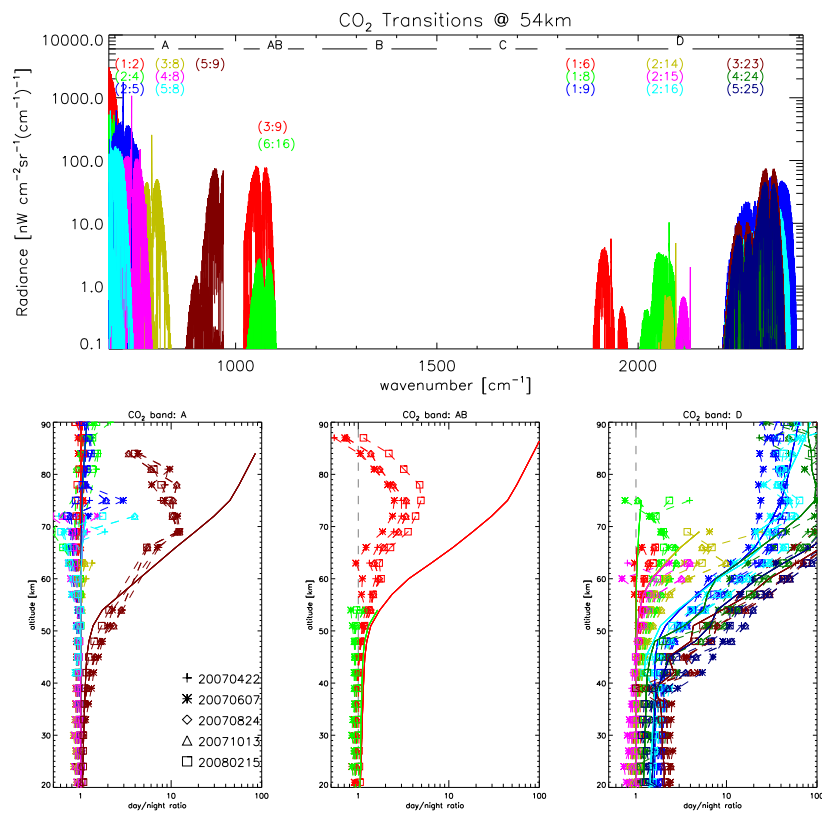


Figure 5: (top) Simulated CO₂ transitions and (bottom) simulated and measured day / night ratio for midlatitude south scenario.

5 Other molecules with no diurnal variation

Inside the MIPAS spectral range the molecules with little or no diurnal variation and which have a reasonable S/N ratio for at least some heights are: H_2O , HNO_3 , CH_4 , N_2O and CO . For these molecules, as in the case of CO_2 , the only difference between the day and night simulated spectra are the vibrational temperatures given by the nonLTE model. In the following subsections, the analysis results correspond to the midlatitude south data but the same conclusions can be drawn from the midlatitude north data.

5.1 H_2O results

In the case of H_2O , its emission spreads over the complete MIPAS spectral range. However, strong “pure” lines required for this analysis appear mainly in the MIPAS bands B, C and D. H_2O emission in these spectral regions corresponds mainly to the transition $\text{H}_2\text{O}(1:2)$. Figure 6 shows that the behaviour of the MIPAS data is consistent from spectral region to spectral region. It can also be noted that the model predicts the observed ratio to within 10%. There is a discrepancy around 40km where the model ratio is greater than one while the measured ratio is always lower than one.

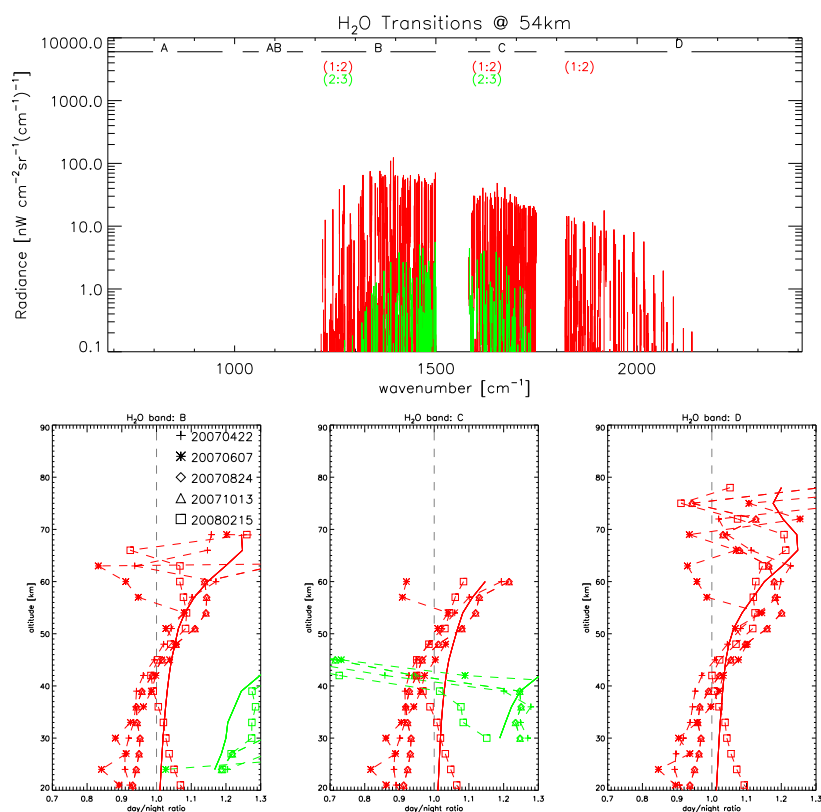


Figure 6: (top) Simulated H_2O transitions and (bottom) day / night ratios as found in the data and in the simulations

5.2 HNO₃ results

Figure 7 displays on the left panel the transitions identified for HNO₃. These transitions are shown for a lower altitude than previously because there are no transitions of HNO₃ above 10% of the NESR expected for the MIPAS reduce resolution measurements above 50km.

For this molecule strong “pure” lines required for this analysis appear only in MIPAS band A. The right panel shows the day/night ratio results. The analysis can only be extended up to about 40km because at higher levels the S/N ratio is poor. At this low levels, thermal collisions are assumed to be frequent enough to maintain the emission in LTE and therefore, the simulated ratio is equal to one. However, the measured ratio seems to be less than one at about 35km and abruptly more than one around 40km. This behaviour is found in the three transitions analyzed in practically all the days.

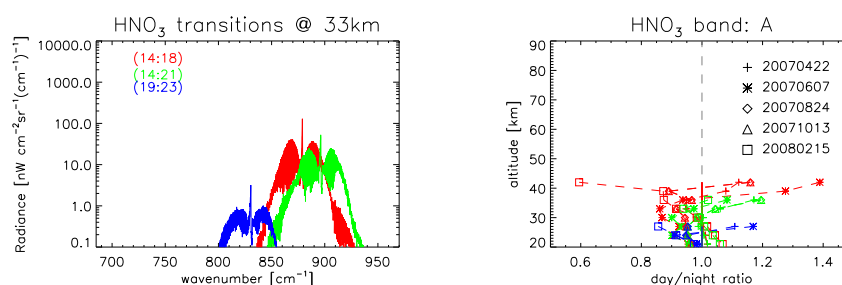


Figure 7: (left) Simulated HNO₃ transitions and (right) day / night ratios as found in the data and in the simulations

5.3 CH₄ results

Figure 8 displays on the left panel the transitions identified for CH₄. For this molecule strong “pure” lines required for this analysis appear only in MIPAS band B. The right panel shows the day/night ratio results. As in the H₂O day/night ratio there is an anomaly around 40km where the model ratio is greater than one while the measured ratio is always lower than one. Above this altitude, the simulated ratio falls within the variability encountered in the measured ratio.

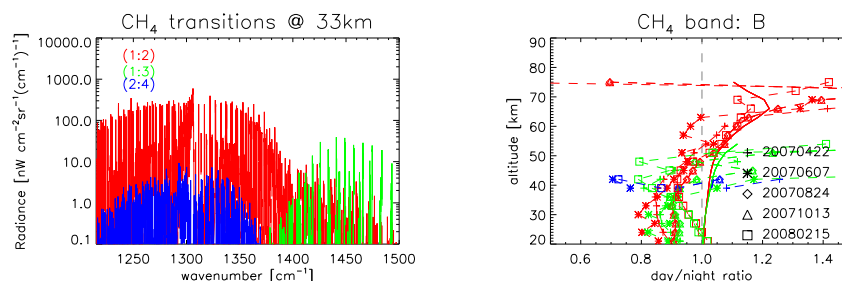


Figure 8: (left) Simulated CH₄ transitions and (right) day / night ratio as found in the data and in the simulations

5.4 N₂O results

Figure 9 displays the N₂O results. In band B, the nonLTE model seems to assume LTE (simulated ratio equal one) while the measured ratio is negative up to 40km and then abruptly becomes positive at around 50km. In band D, the simulated ratio falls inside the huge variability shown in the measured ratio.

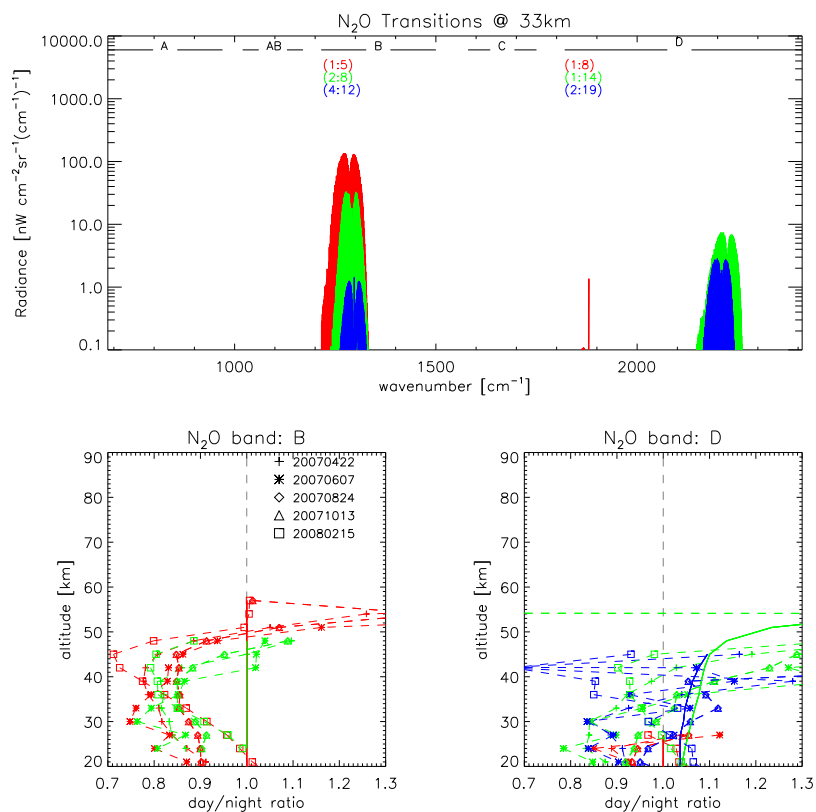


Figure 9: (top) Simulated N₂O transitions and (bottom) day / night ratios as found in the data and in the simulations

5.5 CO results

Figure 10 displays the results for CO. Below 60km, the simulated ratio falls inside the variability encountered inside the measured ratio, however, it seems to lie towards the high values. Above this altitude the simulated ratio always overestimates the measured ratio.

6 Molecules with diurnal variation

The interpretation of the result of this analysis for gases that present a clear diurnal variation is not simple because the ratio will depend of the difference in the day–night concentration as well as in the nonLTE effects. Nevertheless the analysis was applied to NO₂, NO and O₃,

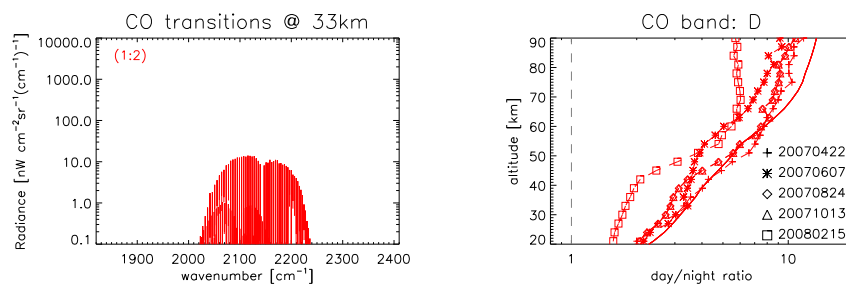


Figure 10: (left) Simulated CO transitions and (right) day / night ratios as found in the data and in the simulations

In the case of NO_2 and NO , the day and night climatological profiles used to compute the simulated spectra include a diurnal variation and therefore the simulated ratio and the measured ratio should show relatively the same behaviour with an offset due to the diurnal variation and the nonLTE differences found in the simulated and in the measured spectra.

In the case of O_3 , the day and night climatological profiles used to compute the simulated spectra are identical, therefore, while the measurement ratio will reflect the nonLTE and concentrations day–night differences, the simulated ratio will only reflect the nonLTE day/night effects predicted by the model.

6.1 NO_2 results

Figure 11 displays on the results for NO_2 . It can be seen that even though this molecule has a strong diurnal variation the simulated ratio shows the same behaviour as the variability encountered in the data. The worst case is around the lowest altitudes where the difference between the simulated ratio and the measured ratio is about 30%. It would be interesting if this difference can be explained by the deviation of the diurnal variability of the NO_2 around these altitude levels, however that is not pursued here.

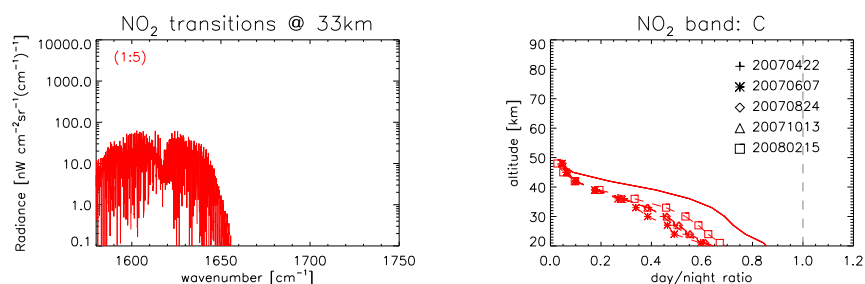


Figure 11: (left) Simulated NO_2 transitions and (right) day / night ratios as found in the data and in the simulations

6.2 NO results

Figure 12 displays the results for NO. Again, as for the NO₂ day/night ratio results, the simulated ratio shows the same behaviour as the measured ratio, however, for this molecule the offset encountered between the measured ratio and the simulated ratio is huge.

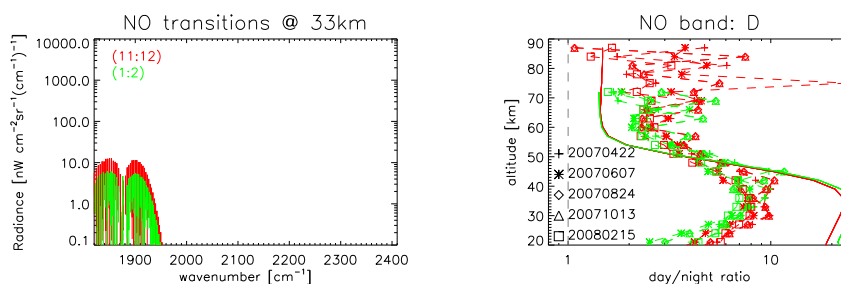


Figure 12: (left) Simulated NO transitions and (right) day / night ratios as found in the data and in the simulations

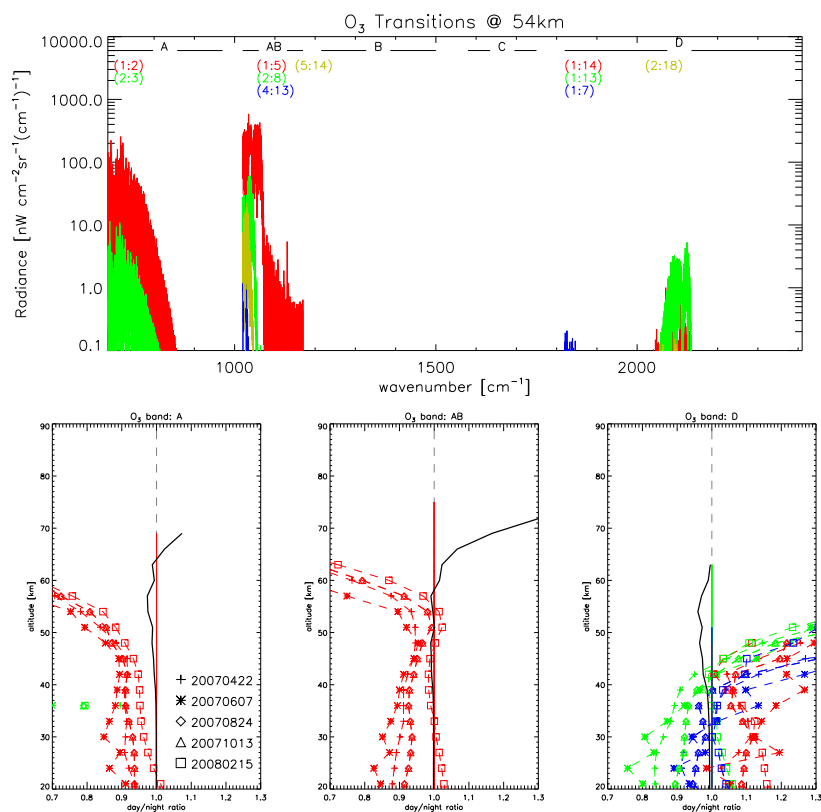


Figure 13: (top) Simulated O₃ transitions and (bottom) day / night ratios as found in the data and in the simulations. The ratio between the day nonLTE and the day LTE is shown in black

6.3 O₃ results

Figure 13 displays the results for O₃. As can be seen, the simulated transitional ratio predicts no diurnal variation in the nonLTE effects. The measured ratio in band A and AB shows clear signatures of the diurnal variation concentration with less ozone during daytime than during nighttime due to its destruction by UV solar radiation. However, the measured ratio in band D shows the opposite behaviour suggesting that some nonLTE effects are present. These nonLTE effects are not reflected in the simulated data and therefore assumed to be not modelled. Figure 13 also shows the ratio (black line) of the day nonLTE and the day LTE simulated radiance to show that there are some nonLTE effects predicted by the nonLTE model. This ratio was calculated with the entire signature of the molecule rather than for a particular transition. The same ratio is found when analyzing the night nonLTE and LTE simulated radiances. Since there is no nonLTE or gas concentration diurnal variation in these simulations, it is difficult to draw conclusions with this technique and further work needs to be carried out to validate the O₃ nonLTE modelling.

7 Summary and conclusions

A method to test the nonLTE model results is introduced. This method was used to analyze the main transitions of gases with no diurnal variation as CO₂, H₂O, HNO₃, CH₄, N₂O and CO, as well as gases with diurnal variation such as, NO₂, NO and O₃. In order to allow for the effect of some SZA variation different days were analyzed.

The analysis of CO₂ transitions inside the MIPAS spectral range revealed a discrepancy in the modelling of the upper level “9” (00⁰1) of the HITRAN global quantum indices, encountered in two different transitions, (5:9) in band A and (3:9) in band AB. The rest of the CO₂ transitions seems to fall inside the atmospheric variability encountered in the measured ratio.

The analysis of H₂O vibrational transition (1:2) revealed that the MIPAS data is consistent between spectral regions. It also displays a discrepancy around 40km where the simulated ratio is always greater than one and the measured ratio is always lower than one. The same kind of discrepancy was found in the analysis of HNO₃, CH₄ and N₂O.

The results of NO₂ and NO prove that even though the ratio for these molecules is not only dependent on the nonLTE diurnal variation, the measured ratio and the simulated ratio shows relatively the same behaviour offset by the diurnal variations and nonLTE differences between the simulated and the measured radiances.

The analysis of the results from O₃ is difficult because there is no nonLTE diurnal variation in the model while it appears to be one in the measurements.

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