1. INTRODUCTION

Methane (CH₄) is released in the atmosphere by natural processes (e.g. wetlands, termites) as well as by anthropogenic activities (e.g. fossil fuel exploitation, rice agriculture, biomass burning). Due to its high warming potential and its relatively long chemical lifetime (9 years), atmospheric methane plays a major role in the radiative forcing responsible of the greenhouse effect. Methane also affects climate by influencing tropospheric ozone and stratospheric water [1]. The cycle of methane is complex and to understand it requires a complete study of its emissions and its budget of sources and sinks. High quality methane data sets are thus necessary to perform such studies.

Methane vertical distributions as well as total and partial column time series can be retrieved from high-resolution ground-based FTIR spectra, using, e.g., the SFIT-2 algorithm which implements the Optimal Estimation Method of Rodgers [2]. However, although several retrieval approaches characterized by relatively high information content exist, methane retrieval profiles very often present large oscillations in their tropospheric part, which might result partly from inappropriate or inconsistent spectroscopic parameters. Significant improvements on retrieval quality could be therefore reached by using more accurate CH₄ spectroscopic data. The main purpose of this contribution is to test and compare three different sets of CH₄ spectroscopic parameters and to quantify their impact on CH₄ retrieved products as well as on the fitting quality.

2. RETRIEVAL STRATEGY

Table 1 presents the 5 microwindows simultaneously fitted during the retrieval procedure adopted here. These microwindows are set as the jointly adopted by all partners involved in the European HYMN project (www.kmi.eur/hamon/hymn). All FTIR spectra inverted in this study, by using the SFIT-2 3.9 release algorithm, to produce CH₄ columns or vertical profiles are the high-resolution (0.003 cm⁻¹ to 0.005 cm⁻¹) FTIR solar observations recorded during the year 2005 at the International Scientific Station of the Jungfraujoch (ISSJ - 46.5°N, 8.0°E, 3580m asl). Only spectra with solar zenith angle lower than 80° have been analyzed. This correspond to a subset of around 440 FTIR spectra. A priori CH₄ profile and diagonal covariance matrix used during the retrieval procedure were obtained from the mean data set of the HITRAN 2004 database with respect to the two other datasets. As we can observed from Table 2 here below, comparisons of retrieved CH₄ total columns using the standard deviations for CH₄ total and partial columns. (X-HIT04)/HIT04)*100) and corresponding standard deviations for CH₄ total and partial columns.

3. IMPACT ON INFORMATION CONTENT & ERROR BUDGET

No significant difference on information content (i.e. averaging kernel functions (AVK)), their corresponding eigenvectors and eigenvalues, the number of degree of freedom of the signal (DOFS) has been observed when characterizing our CH₄ retrievals successfully performed with the HIT-04, the CF and the FH linelists. AVK averaging kernels (left part of Figure 1) and their corresponding three most significant eigenvectors (middle part of Figure 1) are typical examples of information content results obtained for a solar spectra recorded at mean zenith angle (65°) and high resolution (0.003 cm⁻¹). They show a good sensitivity to methane inversions between the altitude site (3.58 km) and almost 30 km. Eigenvalues also indicate that, in that altitude range, the main contribution to the CH₄ retrieval is always coming from the measurement, rather than from the a priori state. In addition, when considering the whole timeseries analyzed here, the DOFS value is close to 3.05 ± 0.27, whatever the spectroscopy used.

The same conclusion can be drawn when comparing, for each atmospheric layer defined by the AVK functions of Figure 1, individual contributions to the total error of the three most common random error sources (smoothing error, measurement error and model parameters error): indeed, no significant difference has been observed and, in all cases, the corresponding error budget of the retrieved VMRs below 30 km is very similar to the one plotted on the right part of Figure 1.

4. IMPACT ON RETRIEVAL PRODUCTS & QUALITY

As we can observed from Table 2 here below, comparisons of retrieved CH₄ total columns using the HITRAN 2004 database with respect to the two other datasets do not shown important differences; even if these ones are significant and greater than the total error of factoring our methane retrieved total columns. Values reported in Table 2 are mean relative differences over the whole year 2005 computed as \((X-H/\text{HIT}04)*100\%\), with X=CF or FH. Corresponding standard deviations on the mean are also indicated. Relative differences for partial columns corresponding to the atmospheric layers defined in Figure 1 of this paper have been calculated. Once again, significant differences are not observed, but, this time, are lower than total errors affecting corresponding partial columns. Except for the (3.58–7) km layer, the CF linelist always gives partial columns lower than those obtained with HIT-04. The FH linelist always gives partial columns larger than the HIT-04 ones, except for the (17-27) km altitude range. For both CF and FH linelists, major differences with HIT-04 retrieved columns are observed for the (7–17) km layer.

In addition, significant differences and sensitive improvements can be observed when considering CH₄ retrieved VMRF profiles. Figure 2 presents retrieval results for a FTIR spectrum recorded on March 1, 2005, at a solar zenith angle close to 80°. While the CF linelists allow to significantly reduce the magnitude of tropospheric oscillations in the HIT-04 retrieved profile, the FH parameters make them totally disappeared (left part of Figure 2). The right part of Figure 2 shows, for each CH₄ microwindow, corresponding residuals (observed minus calculated spectrum). Grey circles indicate residuals structures associated to methane absorption lines. Improvements reached by using CF or FH linelists are clearly visible. To provide a more complete statistics, Table 3 summarizes, for each microwindow, mean residuals values averaged over a sample of almost 330 spectra.

Table 2 - Mean relative differences (computed as \((X-\text{HIT})/\text{HIT}04)*100\%\)) and corresponding standard deviations for CH₄ retrieved and partial columns.

Table 3 - Mean residuals values (computed over a set of 127 spectra) for each CH₄ microwindow and for the HIT-04, CF and FH spectroscopic linelists. Underlined values give better results for each microwindow. These values suggest that CF and FH methane parameters significantly improve fitting quality without introducing a large bias on CH₄ retrieved total and partial columns (see Table 2).

Figure 2 - Example of CH₄ retrieved profiles (left panel) and fitting residuals (right panel) by using the HIT-04, CF and FH spectroscopic linelists. Significant improvements concerning the magnitude of tropospheric oscillations and methane residuals features (grey circles) are reached.

Figure 1 - Typical averaging kernels (AVK), left frames), eigenvectors (middle frames) and error budget (right frames) characterizing our CH₄ retrieved AVK and eigenvectors. Calculations have been performed for a solar spectra recorded at a solar zenith angle of 65°, with a resolution of 0.003 cm⁻¹. The spectroscopy used is that defined in the HITRAN-2004 linelist. Very similar curves are obtained using CF or FH methane spectroscopic parameters.