Impact of different spectroscopic datasets on CH_4 retrievals from Jungfraujoch FTIR spectra

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1. INTRODUCTION

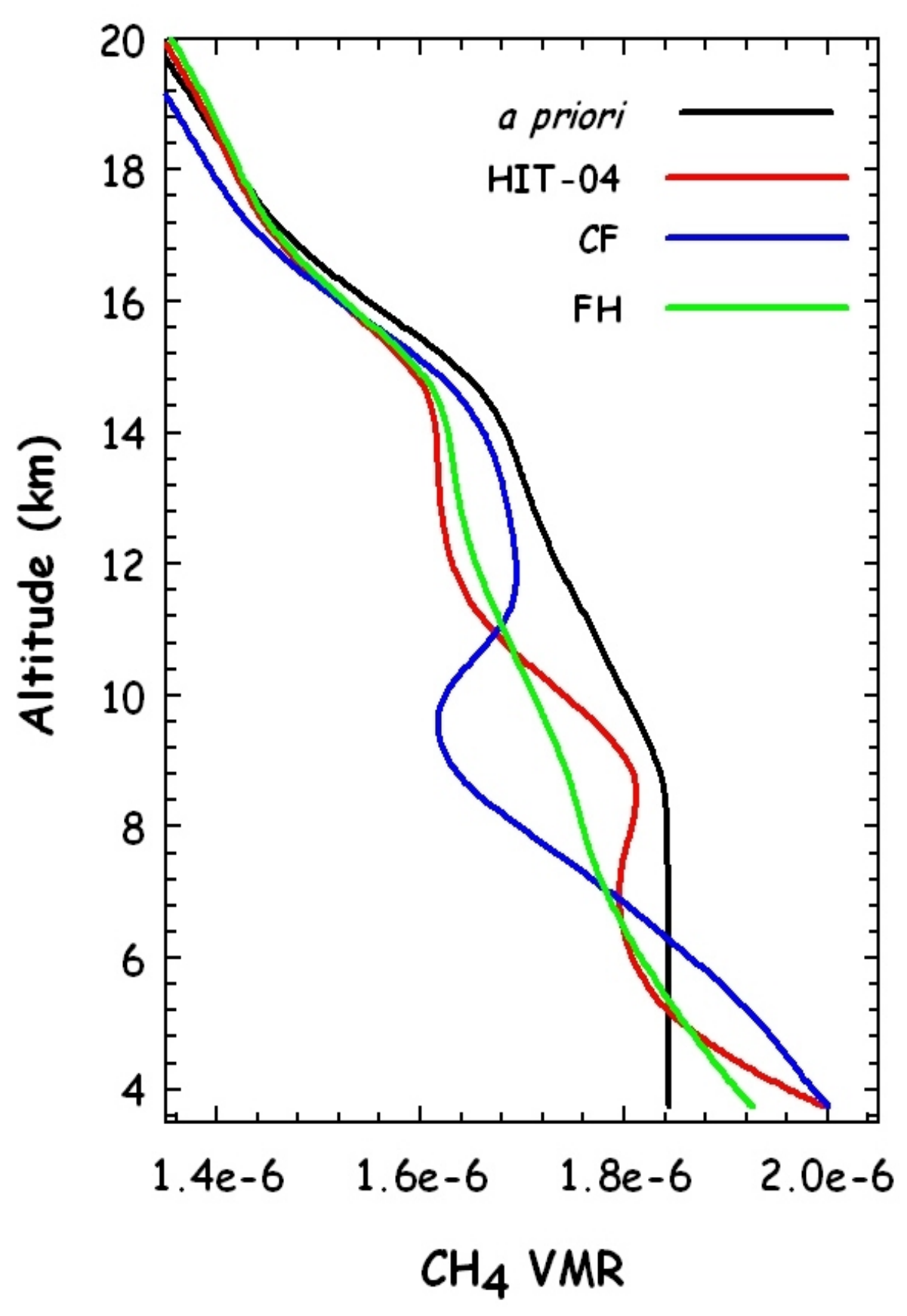
Methane (CH_4) is released in the atmosphere by natural processes (e.g. wetlands, termites) as well as by anthropogenic activities (e.g. fossil fuel exploitation, rice agriculture, biomass burning, etc). Due to its high warming potential and its relatively long chemical lifetime (~9 years), atmospheric 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 retrieved profiles very often present large oscillations in their tropospheric range, which might result partly from inappropriate or inconsistent spectroscopic parameters. Significant improvements on retrieval quality should therefore be reached by using more accurate CH_4 spectroscopic data. The main purpose of this contribution is to test and compare three different sets of CH_4 spectroscopic parameters and to quantify their impact on CH_4 retrieved products as well as on the fitting quality.

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 CH_4 retrievals successively performed with the HIT-04, the CF and the FH linelists. VMR 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 zenithal 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 30km. Eigenvalues also indicate that, in that altitude range, the major contribution to the CH_4 retrievals is always coming from the measurement, rather than from the a priori state. In addition, when considering the whole timeseries analyzed here, the mean 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 affecting the retrieved VMRs below 30 km is very similar to the one plotted on the right part of Figure 1.



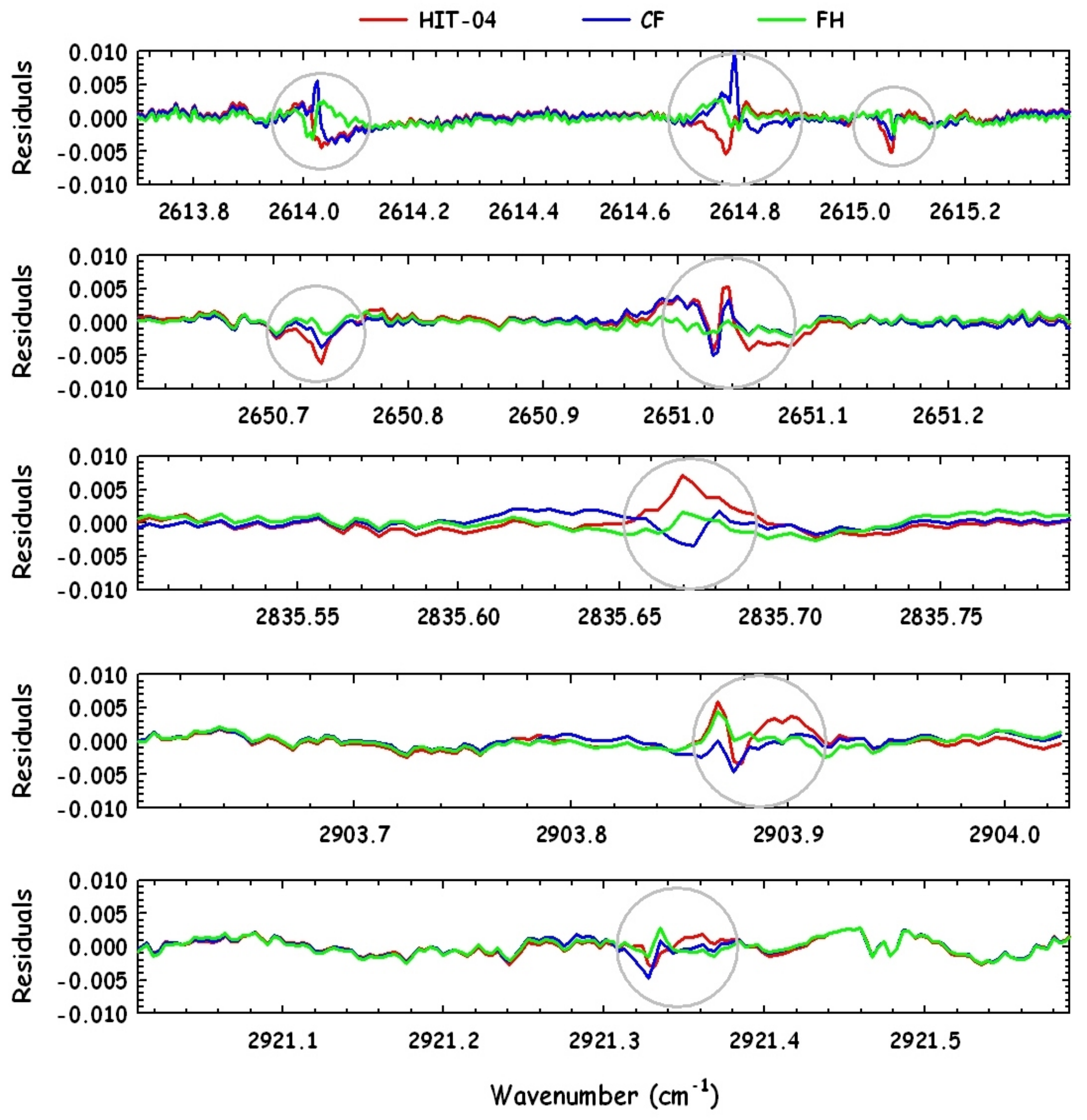


Figure 2 - Example of CH_4 retrieved profiles (left panel) and fitting residuals (right panel) by using the HIT-04, CF and FH spectroscopic linelists. Significant improvements concerning the magnitude of tropospheric oscillations and methane residuals features (grey circles) are reached.

Microwindow	HIT-04	CF	FH
2613	.0992	.0742	<u>.0661</u>
2650	.1032	.0709	<u>.0620</u>
2835	.1195	<u>.0643</u>	.0705
2903	.1339	<u>.0816</u>	.0901
2921	.1673	.1406	<u>.1377</u>

Table 3 - Mean residuals values (computed over a set of 227 spectra) for each CH_4 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 CH_4 retrieved total and partial columns (see Table 2).

2. RETRIEVAL STRATEGY

Table 1 presents the 5 microwindows simultaneously fitted during the retrieval procedure adopted here. This 5 microwindows set is also the one jointly adopted by all partners involved in the European HYMN project (www.knmi.nl/samenw/hymn/). All FTIR spectra inverted in this study - by using the SFIT-2 v.3.91 algorithm which produces CH_4 columns or vertical profiles - are high resolution (0.003 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 about 440 FTIR spectra. A priori CH_4 profile and diagonal covariance matrix used in the retrieval procedure were obtained from zonal mean (for the latitudinal band [41-51]°N) of HALOE space-based measurements. Below 13 km, the a priori CH_4 Volume Mixing Ratio (VMR) profile has however been interpolated downwards to reach CH_4 VMR value close to 1.86 ppm at the altitude site [3].

We have tested three sets of spectroscopic parameters, namely: (i) the original HITRAN 2004 spectroscopic linelist (reported hereafter as "HIT-04") [4]; (ii): two databases where HITRAN 2004 methane line parameters have been updated. These updates are based on recent laboratory measurements performed by C. Frankenberg et al. in the $2\nu_3$ methane band [5]. These laboratory observations have been then fitted independently by C. Frankenberg and by F. Hase, using different fitting tools and procedures, leading to two distinct new CH_4 spectroscopic datasets (reported hereafter as "CF" and "FH", respectively).

Limits (cm^{-1})	Fitted species
2613.70-2615.40	CH_4,CO_2,HDO,solar lines
2650.60-2651.30	CH_4,CO_2,HDO,solar lines
2835.50-2835.80	CH_4
2903.60-2904.03	CH_4,NO_2
2921.00-2921.60	CH_4,H_2O,HDO,NO_2,solar lines

Table 1 - List of microwindows used simultaneously for CH_4 inversions. For each of them, second column provides interfering gases adjusted during the retrieval procedure.

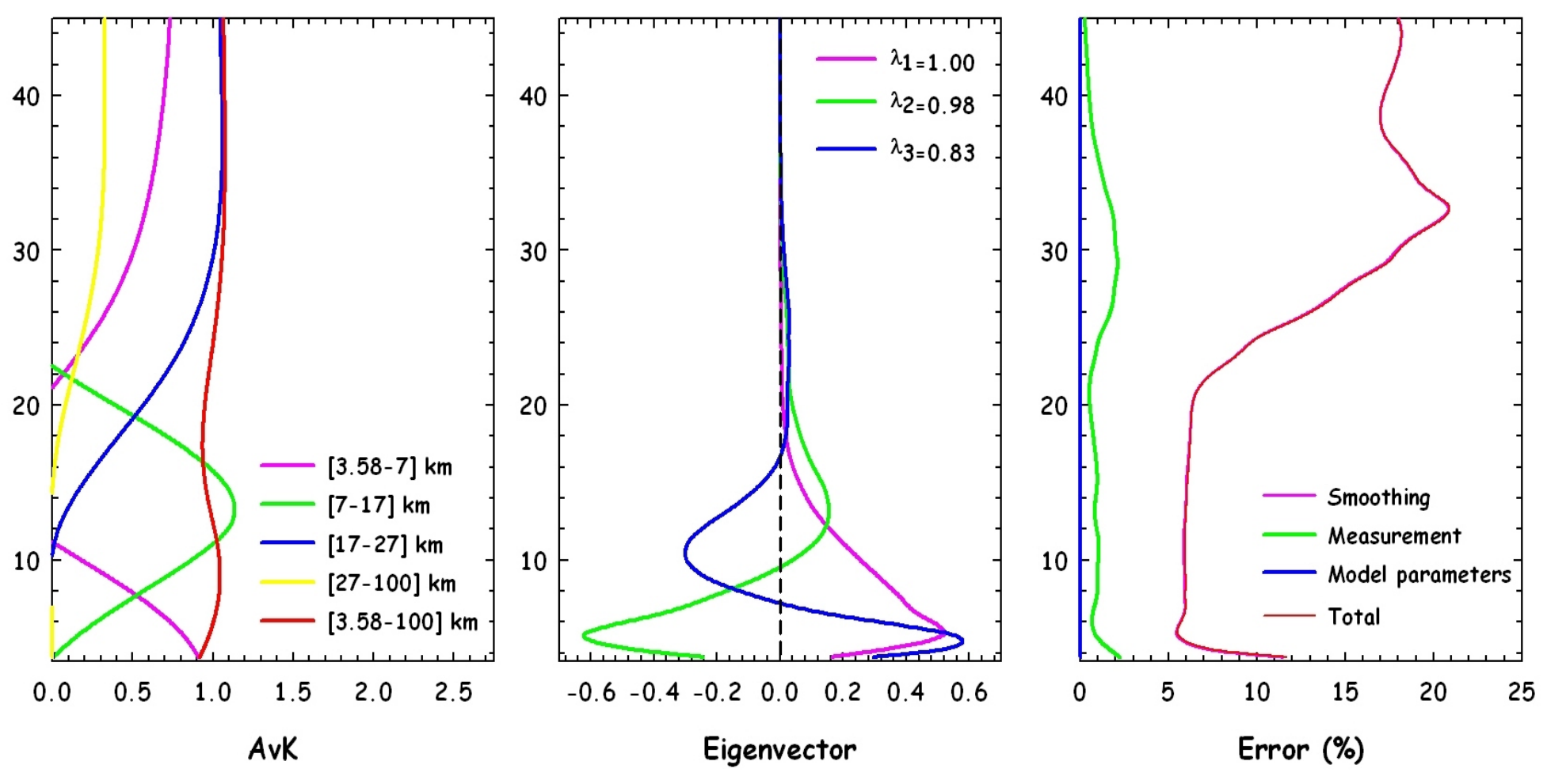


Figure 1 - Typical averaging kernels (AvK, left frame), eigenvectors (middle frame) and error budget (right frame) characterizing our CH_4 retrievals. Calculations have been performed for a spectrum recorded at a solar zenith angle of 65°, with a resolution of 0.003 cm^{-1}. The spectroscopy used is the HITRAN-2004 linelist. Very similar curves are obtained while using CF or FH methane spectroscopic parameters.

4. IMPACT ON RETRIEVAL PRODUCTS & FITTING QUALITY

As can be observed from Table 2 here below, comparisons of retrieved CH_4 total columns using the HITRAN 2004 database with respect to the two other datasets don't shown important differences, even if these ones are significant and greater than the total error affecting our retrieved methane total columns. Values reported in Table 2 are mean relative differences over the whole year 2005 computed as [(X-HIT)/HIT]*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 by AvK of Figure 1 have also been calculated. Once again, bias observed are significant 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 lower than the HIT-04 ones, except for the [17-27] km altitude range. For both CF and FH linelist, 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 CH_4 retrieved VMR profiles. Figure 2 presents retrieval results for a FTIR spectrum recorded on March 1st 2005, at a solar zenith angle close to 80°. While the CF linelist allows to significantly reduce the magnitude of tropospheric oscillations in the HIT-04 retrieved profile, the FH parameters make them totally disappear (left part of Figure 2). The right part of Figure 2 shows, for each CH_4 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.

	X=CF	X=FH
[3.58-100] km	-0.46±0.04	-0.72±0.04
[3.58-7] km	0.85±0.67	-0.63±0.33
[7-17] km	-1.44±0.51	-1.09±0.28
[17-27] km	-0.97±0.61	0.85±0.54

Table 2 - Mean relative differences (computed as [(X-HIT04)/HIT04]*100) and corresponding standard deviations for CH_4 total and partial columns.

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