Approaches for retrieving abundances of methane isotopologues in the frame of the AGACC project from ground-based FTIR observations performed at the Jungfraujoch

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CONTEXT & JUSTIFICATION

This work has been performed within the frame of AGACC (http://www.oma.be/AGACC/Home.html), a contribution to the Belgian Scientific Support for a Sustainable Development. The project intends to make an advanced exploitation of existing ground-based remote-sensing measurements for a selection of atmospheric species that play an important role in the chemistry of the atmosphere and that have a direct or indirect impact on climate. Target species include -among others- lower tropospheric aerosols, H_2O, HDO, CH_4, HCN and CO. The instrumentation includes 3 types of spectrometers (FTIR, MAXDOAS and Brewer) and one CIMEL sun photometer. These instruments are operated at 3 different sites (Jungfraujoch, Ile de la Réunion and Uccle) and most of them are affiliated with the Network for the Detection of Atmospheric Composition Change (NDACC, formerly NDSC - http://www.ndacc.org), an organization dedicated to performing high-quality long-term observations.

Methane 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 (WMO, 2006). The cycle of methane is complex and an extensive study of its main isotopologue, as well as the other isotopic species, is necessary to characterize it. An approach to detect two isotopic varieties of methane ($^{13}CH_4$ and CH_3D) from FTIR measurements is exposed here.

RETRIEVAL STRATEGY FOR $^{13}CH_4$

To study the vertical abundance of $^{13}CH_4$ from high resolution FTIR spectra recorded at the International Scientific Station of the Jungfraujoch (ISSJ) in the Swiss Alps (46.5°N, 8.0°E, 3580m asl - Figure 1), we have selected several $^{13}CH_4$ lines distributed into two sets of microwindows. The first set includes four microwindows in the InSb (2-5.5 µm) detector range, which are fitted simultaneously during the retrievals. The second set contains only one microwindow, in the MCT (7-14 µm) detector range. Characteristics of both microwindows sets are summarized in Table 1.

Range (cm^{-1})	Interfering species	Line #	Lower-state energy E'' (cm^{-1})
InSb range			
2817.38 - 2817.52	none	1	157
2846.15 - 2846.71	$^{12}CH_4$, HDO, H_2O, solar lines	2	63 / 63
2892.45 - 2892.75	$^{12}CH_4$, HDO, H_2O, NO_2	2	63 / 63
2924.15 - 2925.10	$^{12}CH_4$, HDO, H_2O, NO_2, OCS, O_3, solar lines	2	157 / 157
MCT range			
1234.04 - 1234.44	$^{12}CH_4$, $CO^{18}O$, COF_2	1	575

Table 1 - Selection of microwindows for the retrievals of $^{13}CH_4$ in InSb and MCT ranges. For each microwindow, the third column gives the number of $^{13}CH_4$ lines being included while the last column provides, for each line, the corresponding lower-state energy.

For each of these sets, retrievals have been performed using the SFIT-2 v3.91 algorithm and the HITRAN 2004 spectroscopic line parameters database. For the target gas, the retrieved data consist of total or partial column abundances as well as vertical distribution, adjusted from an a priori profile defined on a 41 layers scheme. The adopted a priori $^{13}CH_4$ VMR profile is a zonal mean (for the latitudinal band [41-51]°N), derived from about 400 ACE-FTS space observations performed during the February 2004 - July 2006 time period. The corresponding variability profile has been used to construct the diagonal a priori covariance matrix S_a. For retrieval reasons, only InSb spectra with SZA between 70 and 80° have been analyzed. For the MCT range, the SZA criteria is hardly larger, with zenithal angles between 65 and 80°. Spectral resolutions, defined as the inverse of twice the maximum optical path difference, are 2.85 and 4.96 mK for the InSb domain, 4.00 and 6.10 mK for the MCT range. Under these input and observational conditions, information content analyses give typical degree of freedom (DOF) of 1.0 for the InSb range and of 1.3 for the MCT one. In both cases, it is thus possible to derive only one independent piece of information on the target distribution. This is shown in Figure 2, where the illustrated averaging kernels display a quite good sensitivity from the site altitude up to 14 and 18 km for the InSb and MCT sets, respectively. However, the study of the first eigenvector indicates that sensitivity peaks around 5 km and dramatically decreases below 5 km and above 14 km. Corresponding eigenvalue is typically close to 0.80 for the Insb set and close to 0.90 for the MCT set, meaning that in both cases, the largest fraction of information is coming from the measurement (80 % and 90 %, respectively), rather than from the a priori.

Figure 1 - The International Scientific Station of the Jungfraujoch (ISSJ), in the Swiss Alps (46.5°N, 8°E, 3580m asl), where the University of Liège routinely operates FTIR measurements with 2 high resolution spectrometers. The ISSJ is part of the Network for the Detection of Atmospheric Composition Change (NDACC) primary station.

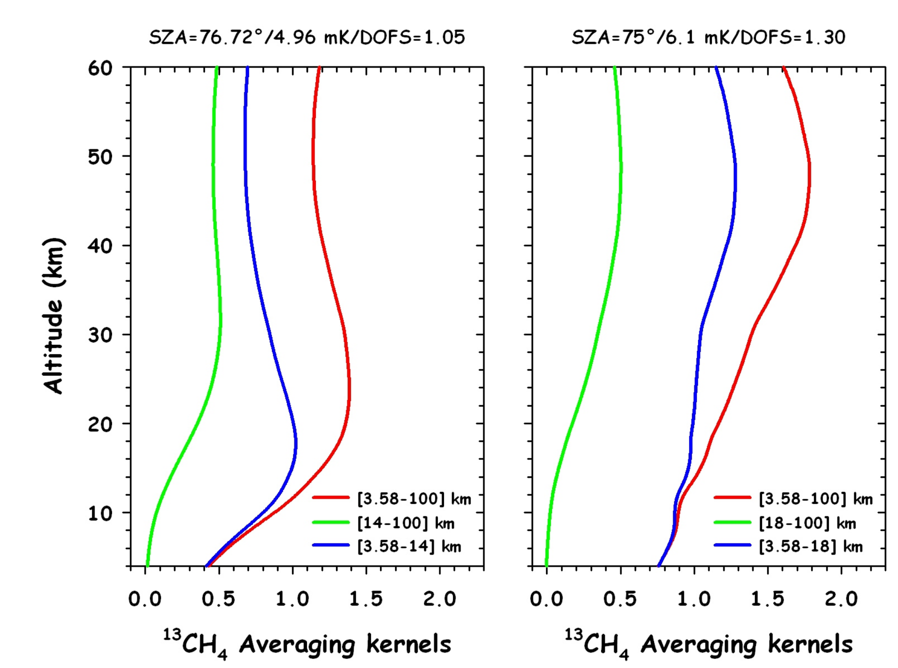

Figure 2 - Typical $^{13}CH_4$ averaging kernels (left: InSb range; right : MCT range) for atmospheric layers defined in the legend. Observationnal conditions adopted for the computation of the kernels are listed above each figure. Degree of freedom of the signal (DOFS) is also given.

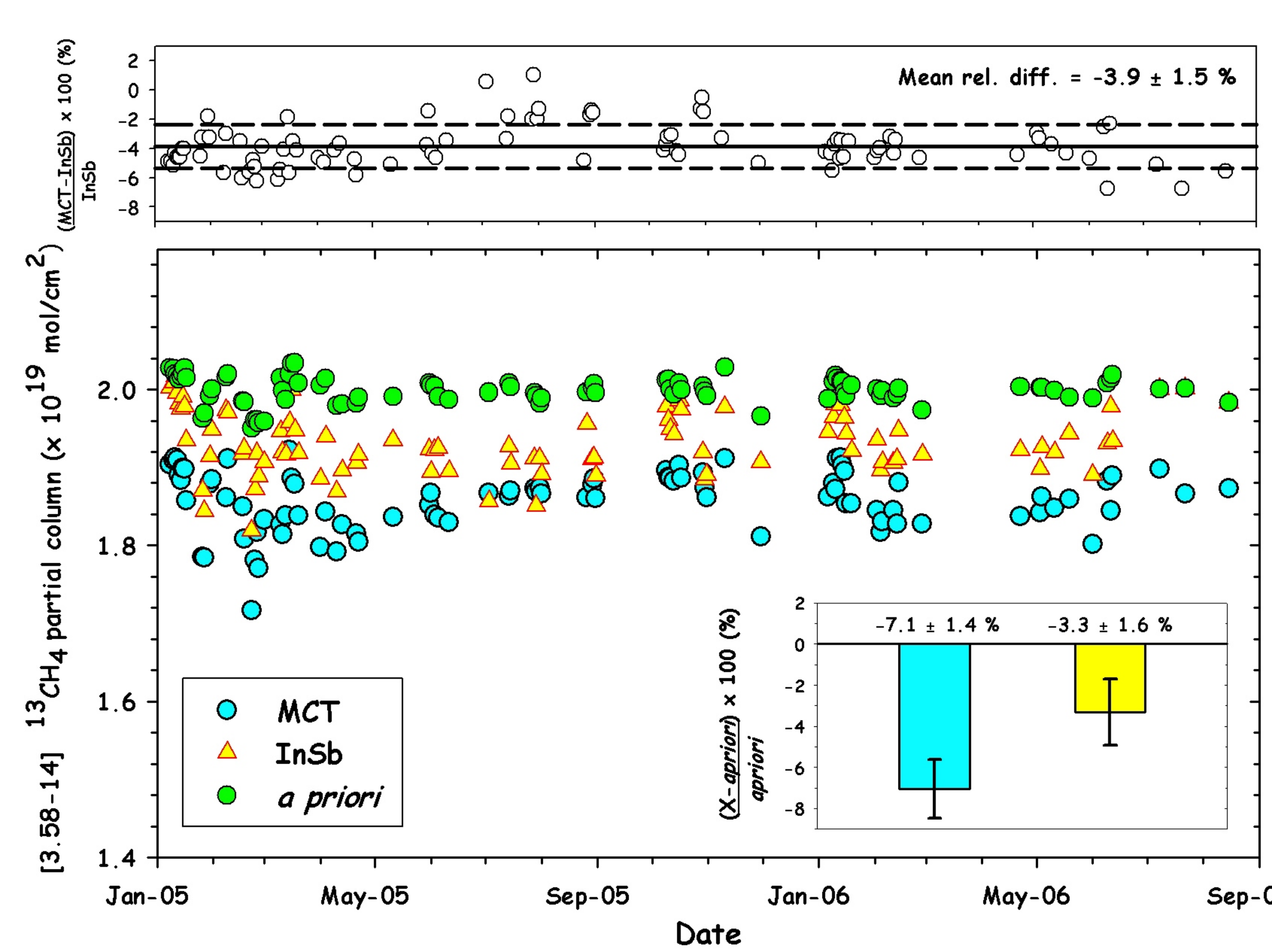

Figure 3 - $^{13}CH_4$ partial columns between 3.58 and 14 km derived from MCT line (light blue dots) and InSb lines (yellow triangles). A priori partial columns are plotted in green. Lower right insert panel : mean relative differences between each retrieved time series and a priori values. Top panel : relative differences between both retrieved time series.

RESULTS FOR $^{13}CH_4$

Figure 2 compares $^{13}CH_4$ partial columns between 3.58 and 14 km, derived from both spectral ranges for the period January 2005 - August 2006. A priori partial columns have also been added to point out the good sensitivity of both microwindow sets selected. The lower right insert panel further gives the relative mean difference, over the whole time period studied, between experimental points retrieved from each spectral range and the a priori values. In the top panel, relative differences between InSb and MCT $^{13}CH_4$ columns reveal a significant mean bias close to 4%. The same mean significant deviation has been observed for the retrieved profiles between 5 and 14 km, indicating that the retrieval strategy doesn't redistribute the $^{13}CH_4$ content above 14 km. However, despite these differences between retrieved values resulting from InSb and MCT ranges, we recommend the use of the MCT microwindow for $^{13}CH_4$ retrievals : the single $^{13}CH_4$ line selected in the MCT range indeed offers a higher information content, a denser set of measurements (due to a wider SZA range for the retrievals) and smaller perturbations by interfering gases (like H_2O and HDO in the InSb set of microwindows).

RETRIEVAL STRATEGY FOR CH_3D

The microwindows selection for the study of CH_3D is not as favorable as for $^{13}CH_4$. A large fraction of the CH_3D lines is indeed polluted by many strong absorptions due to H_2O and HDO. However, we have selected a set of four less-disturbed microwindows, whose characteristics are given in Table 2. Fitting strategy for CH_3D is very similar to the strategy adopted for $^{13}CH_4$. The same a priori VMR and variability profiles have been used for the retrievals performed with v3.91 of the SFIT-2 code. The adopted spectroscopic parameters are also taken from the HITRAN-2004 compilation. Spectral resolutions are 2.85 and 4.96 mK and only FTIR spectra recorded with SZA angles between 70 and 80° have been analyzed.

Range (cm^{-1})	Interfering species	Line #	DOF ($_1$)
2950.77 - 2951.00	HDO, H_2O, O_3	1	0.32 (0.28)
3070.71 - 3071.00	$^{12}CH_4$, O_3	2	0.51 (0.43)
3072.70 - 3073.15	$^{12}CH_4$, H_2O, O_3, solar lines	1	0.51 (0.44)
3089.15 - 3089.70	$^{12}CH_4$, H_2O, O_3	2	0.50 (0.43)
All		6	1.00 (0.76)

Table 2 - Selection of microwindows for the retrievals of CH_3D. For each microwindow, the third column gives the number of CH_3D lines being included while the last column provides the corresponding degree of freedom (DOF). The value between parentheses gives the first eigenvalue ($_1$), indicating the fraction of information coming from the measurement. Last line provides DOF and first eigenvalue for retrievals including all microwindows simultaneously.

Table 2 indicates that a multi-microwindows fitting procedure gives a much higher information content than an individual microwindow fitting procedure. However, at this stage of our CH_3D investigation, it is still unclear as to which fitting strategy is the best one: further tests have to be performed to check if microwindows with large H_2O and HDO absorptions (microwindows #1, #3 and #4) can be included in the retrieval approach for days with high water vapor content and if these wicrowindows are also suitable for lower altitude sites. Other options will also have to be tested like, for example, the feasibility of a multi-spectra procedure including only microwindow #2, which is the less perturbed by H_2O and HDO.

References

World Meteorological Organization, Greenhouse gas bulletin, Bulletin n°2, November 2006.

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