NORS data consistency

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	FUNCTION	NAME	ORGANISATION	DATE	SIGNATURE
LEAD AUTHOR		F. Hendrick	BIRA-IASB	29/10/2013	
CONTRIBUTING AUTHORS		B. Franco	ULg		
		E. Mahieu	ULg		
		M. Van Roozendael	BIRA-IASB		

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I. INTENTION OF THIS DOCUMENT

The aim of this report is to evaluate the consistency of selected NORS data products, namely FTIR and UV-vis DOAS stratospheric NO_2 and H_2CO vertical column densities (VCDs). For this purpose, stratospheric NO_2 and H_2CO VCD retrieved from FTIR and UV-vis DOAS observations at the NORS station of Jungfraujoch have been compared.

II. STRATOSPHERIC NO_2 DATA CONSISTENCY

The comparison between FTIR and UV-vis DOAS stratospheric NO_2 retrievals at Jungfraujoch is extensively described in Hendrick et al. (2012). We provide here the most significant results.

II.1 UV-VIS DOAS STRATOSPHERIC NO_2 RETRIEVAL

A SAOZ (Système d'Analyse par Observation Zénithale) instrument has been operated by BIRA-IASB at the Jungfraujoch station since early 1990's. It is a broad-band (300-600 nm), medium resolution (~1 nm) diode-array UV-vis spectrometer that measures zenith scattered sunlight (Pommereau and Goutail, 1988). Zenith radiance spectra are analyzed using the DOAS (Differential Optical Absorption Spectroscopy) technique (Platt and Stutz, 2008). NO_2 is retrieved in the 425-490 nm wavelength range, taking into account the spectral signatures of NO_2, O_3, H_2O, O_4, and the filling-in of the solar Fraunhofer bands by the Ring effect (Grainger and Ring, 1962). The NO_2 absorption cross-sections at 220 K are from Vandaele et al. (1998). A third-order polynomial is used to fit the low frequency spectral structure due to molecular and Mie scattering.

NO_2 vertical column densities are derived from vertical profiles retrieved by applying an Optimal Estimation Method (OEM)-based profiling technique to sunrise and sunset NO_2 differential slant column densities (DSCDs) which are the direct product of the DOAS analysis. The forward model includes a stacked photochemical box model in order to reproduce the rapid variation of NO_2 at twilight. This model also provides *a priori* profiles to the OEM algorithm and is used to photochemically convert the retrieved profiles, which are representative of twilight conditions, to the mean SZA corresponding to the FTIR observations. NO_2 profile retrievals are quality-checked based on the retrieval fit residual, i.e. RMS of the difference between measured SCDs and those calculated using the retrieved profiles. A detailed error budget can be found in Hendrick et al. (2004).

II.2 FTIR STRATOSPHERIC NO_2 RETRIEVAL

ULg is in charge of the operation of a high-resolution FTIR Bruker (120HR) instrument at the Jungfraujoch station. Solar observations are collected under clear-sky conditions via local or remote control. For this study, all available FTIR observations since 1990 have been fitted with the V3.91 of the OEM-based SFIT-2 algorithm, using pressure and temperature information provided by the National Centers for Environmental Prediction (NCEP, see http://www.ncep.noaa.gov) and assuming the HITRAN 2004 spectroscopic line parameter

compilation, including the August 2006 updates (Rothmann et al., 2004, see also http://www.cfa.harvard.edu/hitran). In the present case, two microwindows ranging from 2914.6 to 2914.7 cm^{-1} and from 2915 to 2915.11 cm^{-1} have been fitted; *a priori* vertical profiles for fitted (H_2O, CH_4 and O_3 scaled during the retrieval process) and simulated interferences (e.g. H_2CO) were based on predictions obtained with the version 5 of the WACCM model (Whole-Atmosphere Community Climate Model, see Chang et al., 2008). For NO_2, 12 monthly profiles based on the same photochemical model as for the SAOZ profile retrieval were used (see Sect. II.1). The total random and systematic errors are of 11% and 36%, respectively.

II.3 INFORMATION CONTENT COMPARISON

The information content associated with both FTIR and SAOZ profile retrievals has been compared. As can be seen in Figure 1, both techniques show similar column averaging kernels and degree of freedom for signal (DOFS) values indicating that they have similar vertical resolution and sensitivity to the vertical distribution of NO_2. Therefore, retrieved FTIR and SAOZ NO_2 columns can be directly compared.

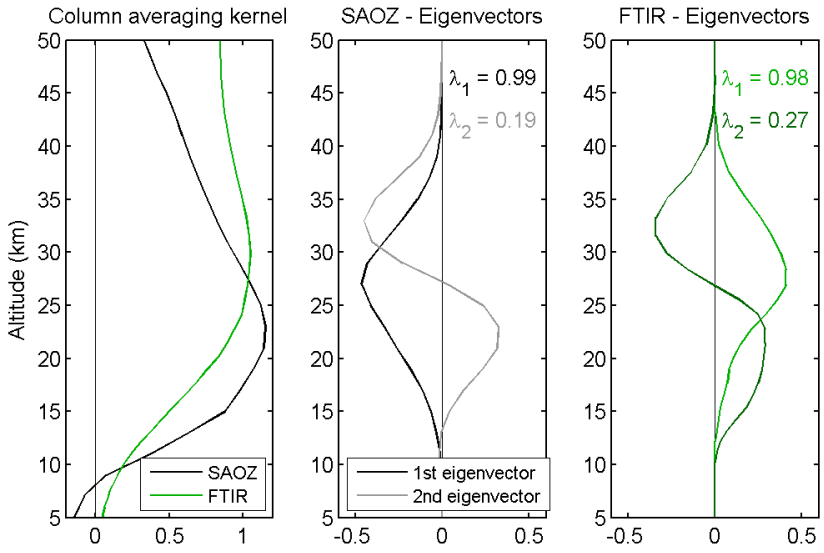

Figure 1: Typical FTIR and SAOZ column averaging kernels (left plot) and leading eigenvectors and corresponding eigenvalues (middle plot: SAOZ; right plot: FTIR). The sum of the latter gives the DOFS.

II.4 FTIR VERSUS UV-VIS DOAS STRATOSPHERIC NO_2 VCD COMPARISON

A very good consistency is found between FTIR and SAOZ data sets, with FTIR measurements lower than SAOZ by 7.8 ± 8.2% on average (see Figure 2). Combining the facts that both retrievals used similar *a priori* profiles and have similar sensitivity to the vertical distribution of NO_2, the remaining differences between FTIR and SAOZ NO_2 columns are consistent with the uncertainties affecting the respective spectroscopic parameters.

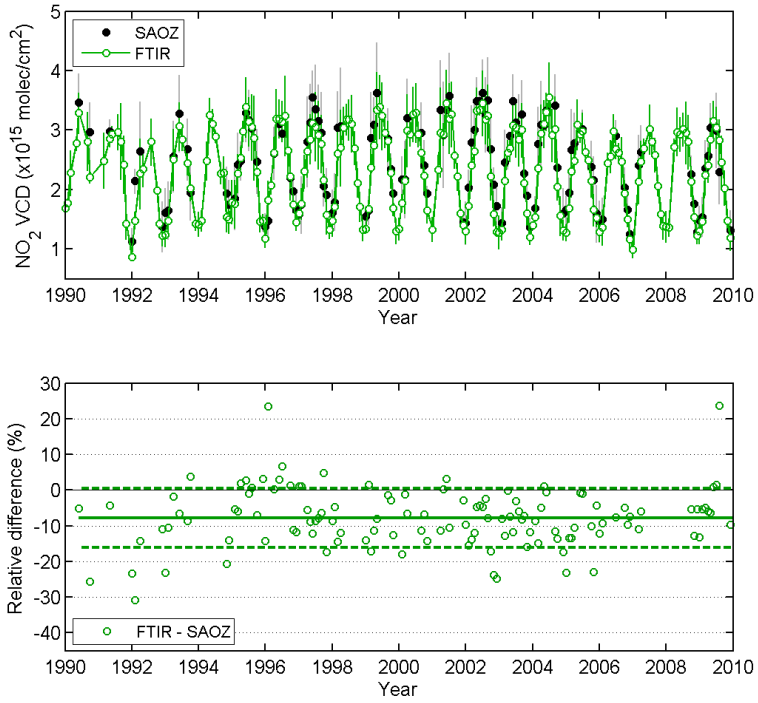

Figure 2: Comparison between FTIR and SAOZ monthly mean stratospheric NO_2 columns at Jungfraujoch (46.5°N, 8°E) for the 1990-2009 period. To ensure photochemical matching, i.e. comparison in the same photochemical conditions, the SAOZ columns, representative of twilight conditions, are converted on a daily basis to the mean FTIR measurement SZA using the BIRA-IASB stacked box photochemical model PSCBOX. The error bars correspond to the 1-sigma standard deviation (natural variability). The relative differences appear on the lower plot with solid and dashed green lines corresponding to the mean FTIR – SAOZ difference and its 1-sigma standard deviation, respectively, which is -7.8 ± 8.2%.

III. H_2CO DATA CONSISTENCY

Similarly to stratospheric NO_2, ULg and BIRA-IASB have started a comparison study between FTIR and UV-vis DOAS retrievals of formaldehyde (H_2CO) at Jungfraujoch. The most significant preliminary results are presented here.

III.1 UV-VIS DOAS H_2CO RETRIEVAL

A MAX-DOAS instrument has been operating by BIRA-IASB at the Jungfraujoch station since July 2010. It is a dual-channel system composed of two grating spectrometers covering the UV and visible wavelength ranges (300-390 nm and 400-720 nm, respectively). A full MAX-DOAS scan requires ~20 min, comprising the following 17 elevation angles: -10°, -8°, -6°, -4°, -2°, 0°, 1°, 2°, 3°, 4°, 5°, 8°, 10°, 12°, 15°, 30°, and 90° (zenith). Only the 0° and positive elevation angles are exploited here. The measured scattered light spectra are analyzed using the settings for H_2CO published by Pinardi et al. (2013). In brief, H_2CO is retrieved in the 336.5-359 nm wavelength range, taking into account the spectral signatures of H_2CO, NO_2, O_3, BrO, O_4, and the Ring effect. The H_2CO absorption cross-sections at 293K are from Meller and Moortgat (2000). A third-order polynomial is used to fit the low frequency spectral structure due to molecular and Mie scattering.

H_2CO vertical profiles and corresponding VCDs have been retrieved from measured H_2CO slant columns by applying the OEM-based bePRO inversion algorithm developed at BIRA-IASB (Clémer et al., 2010; see also Hendrick et al., 2013). This algorithm includes a two-step approach: First, the aerosol extinction vertical profiles are retrieved at 360 nm for each MAX-DOAS scan from the corresponding measured O_4 slant columns. In the second step, the bePRO algorithm is applied to the measured H_2CO slant columns using the retrieved aerosol extinction profile as input. In the latter step, exponentially decreasing a priori profiles with a fixed scaling height of 0.5 km were chosen. As for stratospheric NO_2, H_2CO profile retrievals are quality-checked based on the retrieval fit residual (RMS of the difference between measured SCDs and those calculated using the retrieved profiles). A second approach based on the geometrical approximation has been also used to evaluate H_2CO VCDs. In this method, it is assumed that the H_2CO layer is located below the scattering altitude at 30° elevation, so that tropospheric H_2CO VCDs can be derived by applying a geometrical AMF to measured slant columns at 30° elevation (Hönninger et al., 2004; Brinksma et al., 2008; see also Hendrick et al., 2013). In the preliminary comparison with FTIR data, only the geometrical approach will be involved. The reason is that the selection of good OEM retrievals using the RMS-based selection criterion causes the rejection of a large number of scans in winter. This issue is currently under investigation.

III.2 FTIR H_2CO RETRIEVAL

The Jungfraujoch data set investigated in this study includes 2343 solar spectra recorded at high spectral resolution (0.00513 cm^{-1}) with the Bruker spectrometer, covering the whole December 2005 – April 2013 period. Due to the very weak solar absorption of H_2CO in the IR domain, in particular at the high altitude unpolluted Jungfraujoch site (generally well below 1%, as shown in Figure 3), we set up a tunable narrow bandpass optical filter since December 2005. Using a narrow spectral range (from 2810 to 2850 cm^{-1}) and a larger diaphragm aperture (1.45 mm), this tunable optical filter aims at enhancing the signal-to-noise (S/N) ratio and makes the recorded solar spectra more exploitable for the H_2CO retrieval. Therefore it has been decided to focus first on the 2833.070 – 2833.350 cm^{-1} microwindow only (which absorptions are simulated in Figure 3), conversely to Vigouroux et al. (2009) who used 6 other microwindows ranging from 2763.425 to 2856.400 cm^{-1} (not covered by the tunable optical filter).

All the FTIR observations have been fitted with the SFIT-2 v.3.91 retrieval algorithm, assuming the HITRAN 2004 (including the August 2006 updates) spectroscopic line parameter compilation for the interfering species (HDO, CH_4, O_3 and N_2O, simulated in Figure 3) and HITRAN 2008 for H_2CO only. Pressure and temperature profiles at Jungfraujoch are based on NCEP data. The H_2CO vertical profile simulated by the WACCM v.6 model above Jungfraujoch has been selected as *a priori* for the forward model due to its good agreement with ACE-FTS v.3 occultation measurements as well as with simulations from the IMAGES and GEOS-CHEM models (as shown in Figure 4). *A priori* vertical profiles for the interfering species are also provided by predictions from the WACCM v.6 model.

The H_2CO inversion is performed with the SFIT-2 algorithm using a Tikhonov type L_1 regularization, for which the regularization strength (α parameter) has been fixed at a value of

250. Investigations of various α revealed this value to be a good compromise between percentages of unphysical retrieved profiles on the one hand, and retrieved variables on the other hand. Indeed, α values lower than 250 resulted in increasing percentages of profile with negative volume mixing ratios (VMRs), while α values higher than 250 led to retrieved variables such as total columns, global residuals or degree of freedom for signal (DOFS), getting closer to the results obtained by a simple scaling of H_2CO.

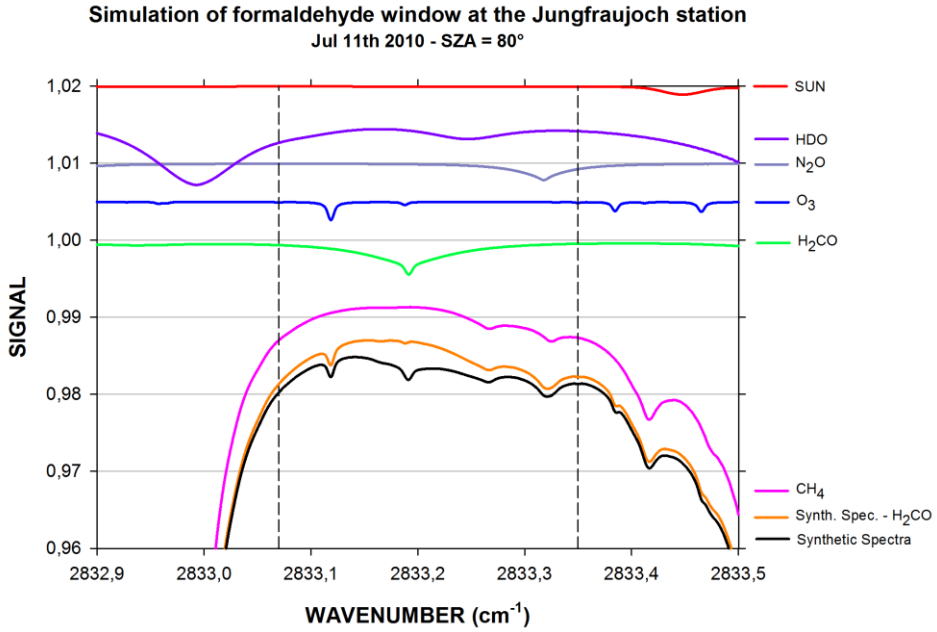

Figure 3: Simulation of the main absorptions in the 2833.070 – 2833.350 cm^{-1} microwindow delimited by the dashed black lines, for a solar zenith angle (SZA) of 80° and under summer conditions. The solid black line is the synthetic spectrum representing the total absorption, and the solid orange line corresponds to the synthetic spectrum without taking into account the H_2CO absorption. This figure reveals the weak absorption and the broad spectral line of H_2CO.

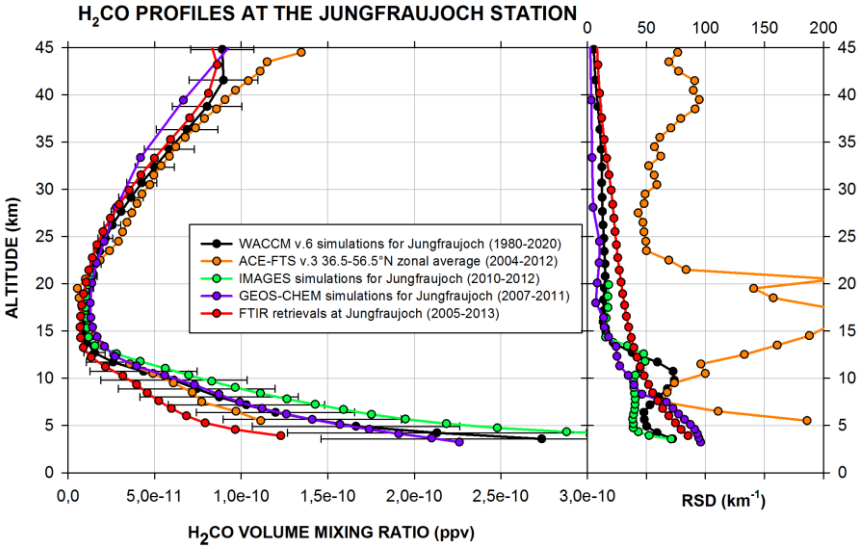

Figure 4: The left chart shows the mean vertical profiles of H_2CO (in VMRs) above Jungfraujoch, simulated by the WACCM v.6 (in black), IMAGES (in green) and GEOS-CHEM (in purple) models and derived from ACE-FTS v.3 occultation measurements (in orange). The average profile produced by the FTIR retrievals is drawn in red. The relative standard deviations (RSD) integrated per vertical km are presented in the right chart.

Figure 5 presents an example of the improvement brought by the fitting of H_2CO solar spectra with the Tikhonov regularization compared to the results obtained by a simple scaling. While the latter is unable to reproduce the entire H_2CO feature around 2833.18 cm^{-1}, hence inducing large residuals, the Tikhonov regularization allows for a better fit of this feature and helps to reduce the residuals significantly near this spectral line (from -0.0004 to -0.0002 in this example). Nevertheless, strong discrepancies between observed and retrieved spectra (in blue and red in Figure 5, respectively) remain on the right side of the H_2CO feature for both the scaling and the fitting approaches.

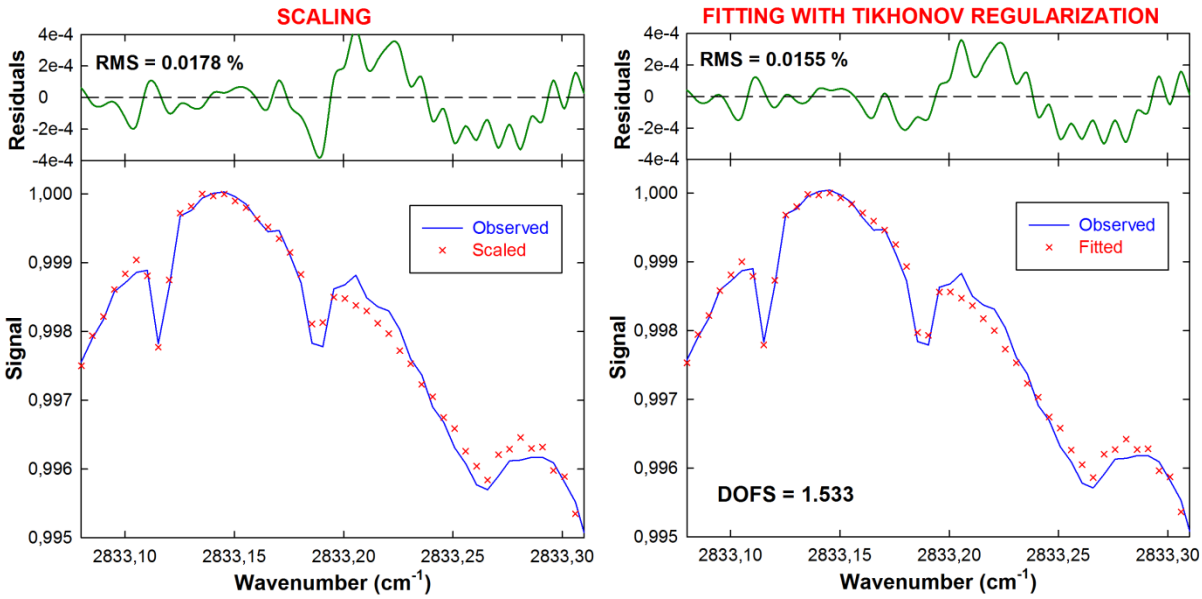

Figure 5: Comparison between observed and scaled/fitted spectra (in solid blue line and red plots, respectively), with the related residuals (in green), for FTIR observations made at the Jungfraujoch station on the 23rd June 2010, for a SZA of 77°. On the left are the results obtained by a simple scaling, while the right chart presents a fitting performed with a Tikhonov regularization (α value fixed at 250).

III.3 PRELIMINARY FTIR VERSUS UV-VIS DOAS H_2CO VCD COMPARISON

The mean seasonal cycles of the total H_2CO column above Jungfraujoch as deduced from the FTIR, MAX-DOAS (geometrical approximation) but also ACE-FTS v.3 data sets are presented in Figure 6. Moreover, ground-based and satellite observations are also compared to partial columns above 3.5km produced by IMAGES 2.5° x 2.5° and GEOS-CHEM 4° x 5° simulations.

From Figure 6, we see that observed and modeled H_2CO total columns show a good consistency in terms of seasonal cycle with a maximum in summer and a minimum in winter. The comparison also reveals systematically lower FTIR H_2CO abundances throughout the year but especially in summer, leading therefore to a lower seasonal amplitude compared to the other data sets. However, it should be noted that a reasonably good agreement is found

between FTIR and MAX-DOAS in January-April and October-December periods. Regarding IMAGES and GEOS-CHEM model data, they appear to be systematically larger than the ground-based and satellite observations, especially during summertime. Investigations are currently under progress in order to better characterize the remaining discrepancies between the different data sets. In the last part of the NORS project, we will focus on the improvements of FTIR and MAX-DOAS retrievals and on the smoothing of the model profiles with ground-based averaging kernels.

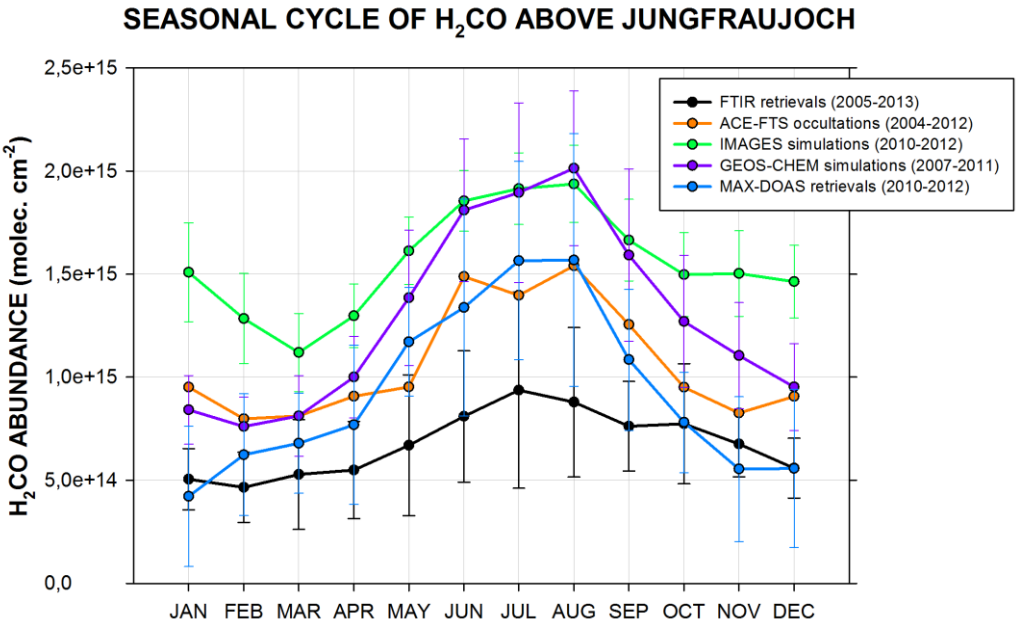

Figure 6: Mean seasonal cycle of H_2CO abundance (expressed in molec. cm^{-2}) above Jungfraujoch with standard deviations, deduced from FTIR retrievals (in black), IMAGES and GEOS-CHEM simulations (in green and purple, respectively), ACE-FTS v.3 occultation measurements (in orange) and MAX-DOAS data (in light blue). It should be noted that ACE-FTS v.3 satellite data consist in partial columns above 6.5km combined with a constant 3.5 – 6.5km partial columns integrated from WACCM v.6 H_2CO simulations.

IV. CONCLUSIONS

- Good agreement between FTIR and UV-VIS DOAS stratospheric NO_2 retrievals at Jungfraujoch with FTIR measurements lower than SAOZ by 7.8 ± 8.2% on average. Both techniques are also very consistent in terms of information content.

- Preliminary comparison between FTIR and UV-VIS DOAS H_2CO total column retrievals at Jungfraujoch shows a good agreement in winter but not in summer where FTIR significantly underestimates UV-VIS DOAS data. It should be also noted that models significantly overestimate FTIR and UV-VIS DOAS observations in summer. These issues will be further investigated in the last part of the NORS project.

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