Use of GEOS-Chem for the interpretation of long-term FTIR measurements at the Jungfraujoch and other NDACC sites

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RECENT INCREASE OF ETHANE ABOVE NORTH AMERICA
(Bruno Franco et al.)

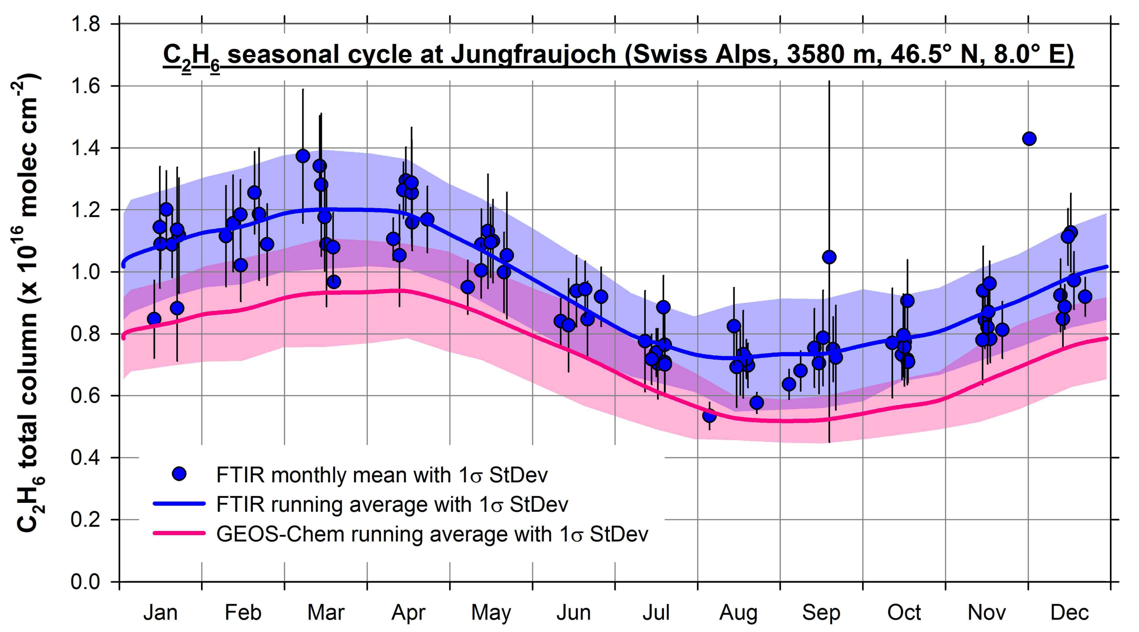


FIGURE 3 - Seasonal variation of ethane as measured at the Jungfraujoch station (in blue) and modeled by GEOS-Chem (v9-2; in red) for the days of observations over the mid-2005 - mid-2013 time period. We have taken into account the vertical resolution and specific sensitivity of the FTIR retrievals before comparison with the model data. Although the seasonal signal is well captured by GEOS-Chem, we observe a systematic bias with an underestimation of the atmospheric amount of ethane by the model. The two data sets cannot be reconciled by accounting for the systematic errors affecting the observations. For more details, see Franco et al., J. Quant. Spectrosc. Radiat. Transfer, 160, 36-49, 2015 (http://hdl.handle.net/2268/175442).

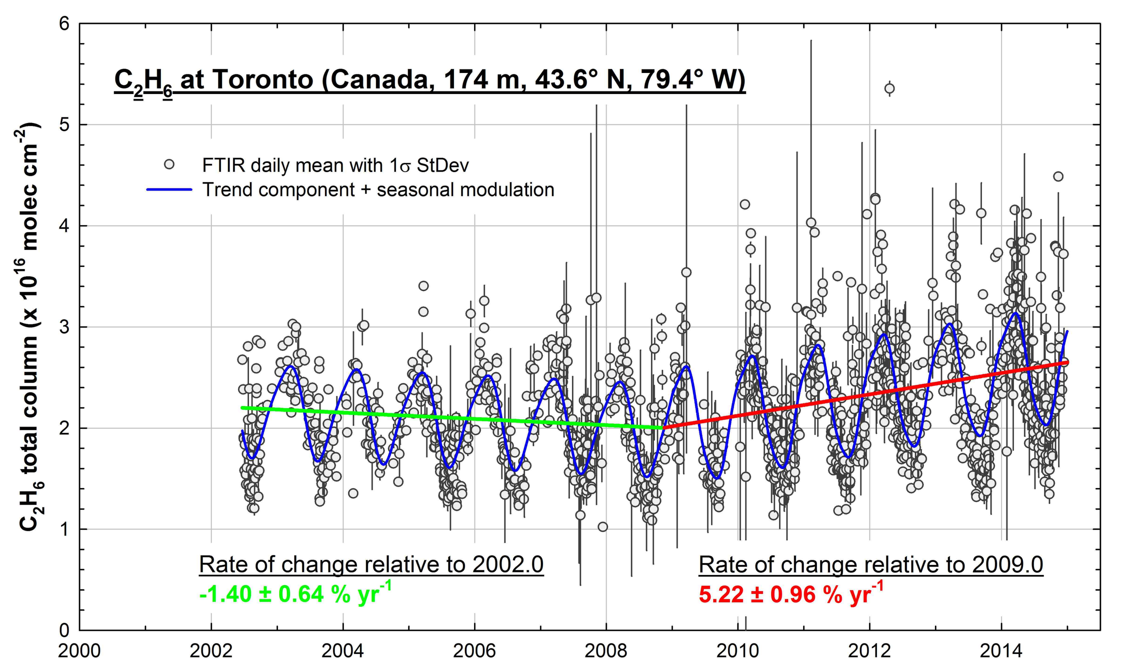
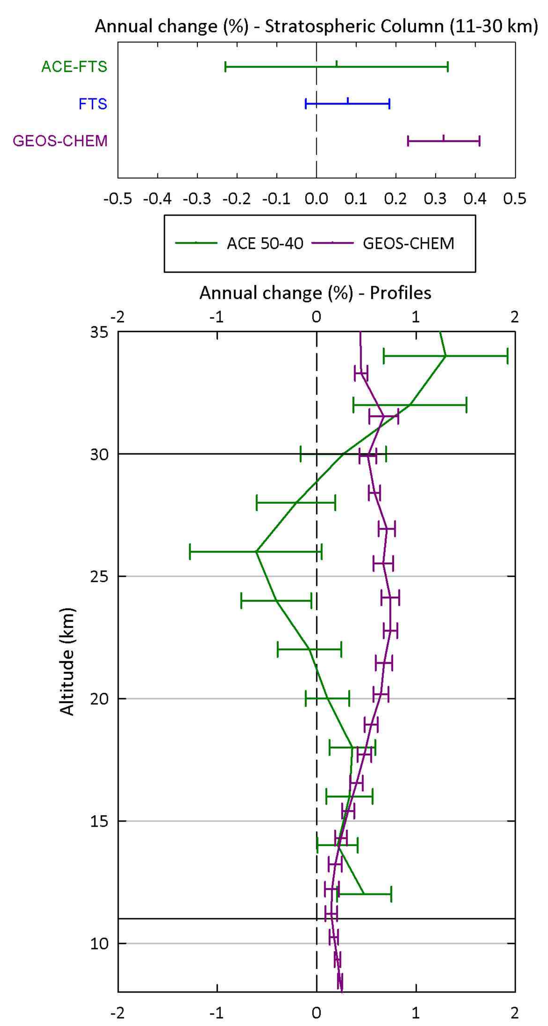


FIGURE 4 - Trends of ethane for Toronto, as deduced from long-term FTIR monitoring activities performed within the framework of the NDACC network. Consistent trends are observed at other sites (e.g. Boulder, CO) and confirmed by ACE-FTS occultation measurements above North America. The recent and massive growth in the exploitation of shale gas and tight oil reservoirs is a candidate explanation for the significant C_2H_6 increase as of 2009 above North America, and more generally in the Northern Hemisphere. Efforts are ongoing to update the emission inventories implemented in GEOS-Chem and evaluate the magnitude of the fugitive emissions required to reconcile the observed and simulated time series of ethane and to assess their impact on air quality. For more details, see Franco et al., EGU2015-4675, 2015 (http://hdl.handle.net/2268/180485).

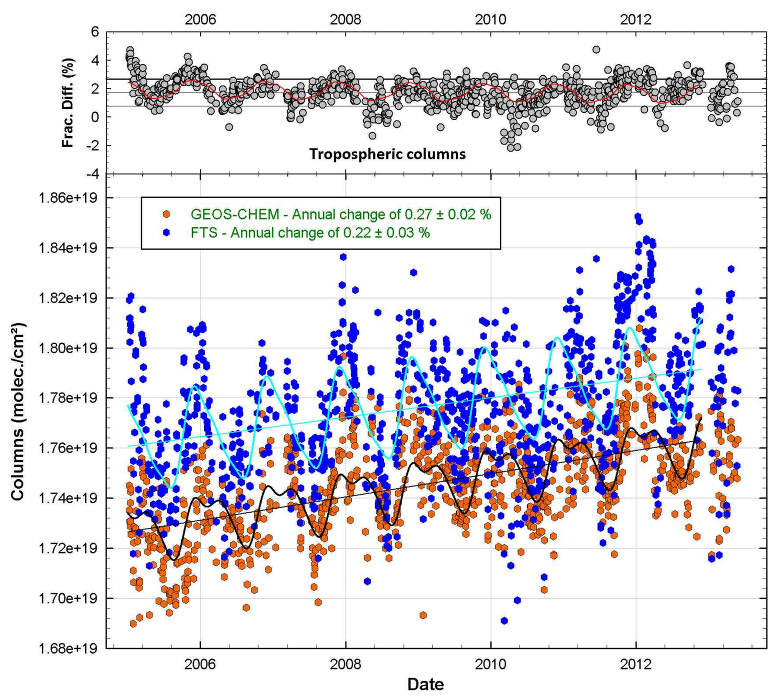
INVESTIGATING THE CAUSES FOR THE METHANE RISE AFTER 2005
(Whitney Bader et al.)



The attribution of the CH_4 increase since 2005 to any source is difficult since the existing measurements datasets (FTIR, *in situ*, satellite …) are insufficient to characterize emissions by region and source process, emphasizing the need for source-tagged model simulations implementing reliable emission schemes. This study focuses on the analysis of the GEOS-Chem CH_4 tagged simulation for six NDACC stations: Eureka, Toronto, Jungfraujoch, Tsukuba, Lauder and Arrival Heights. It should provide information on processes causing the increase of atmospheric methane, provided that we determine consistent trends between the observations and the simulations at the various sites.

FIGURE 6 - A vertical bias between FTIR measurements and the GEOS-Chem simulation has been identified. It stands out that the annual changes of methane in the troposphere (3.58 - 11.7 km) computed from our measurements and GEOS-Chem (v9-2) simulation are in agreement, contrarily to the changes in the total and stratospheric (11.7 – 30.7 km) columns. Comparisons of the annual change of stratospheric CH_4 from our FTS at Jungfraujoch with ACE-FTS measurements (occultations between 50 and 40°N, see figure attached) along with the GEOS-Chem simulation shows an overestimation of the annual change of methane between 11 and 30 km by the model.

FIGURE 7 - Daily mean total column time series for the FTIR measurements (dark blue) and the GEOS-Chem simulation (in grey) are reproduced on the main frame, for the 961 available coincidences over the 2005-2013 (May) time period. Trends computed with a bootstrap resampling tool (Gardiner et al., Atmos. Chem. Phys., 8, 2008) are shown, they agree within the associated uncertainty (2-sigma). The upper frame displays fractional differences {(FTIR – GC)/((FTIR+GC)/2)}, in %.



RETRIEVAL OF FORMALDEHYDE FROM AN UNPOLLUTED SITE
(Bruno Franco et al.)

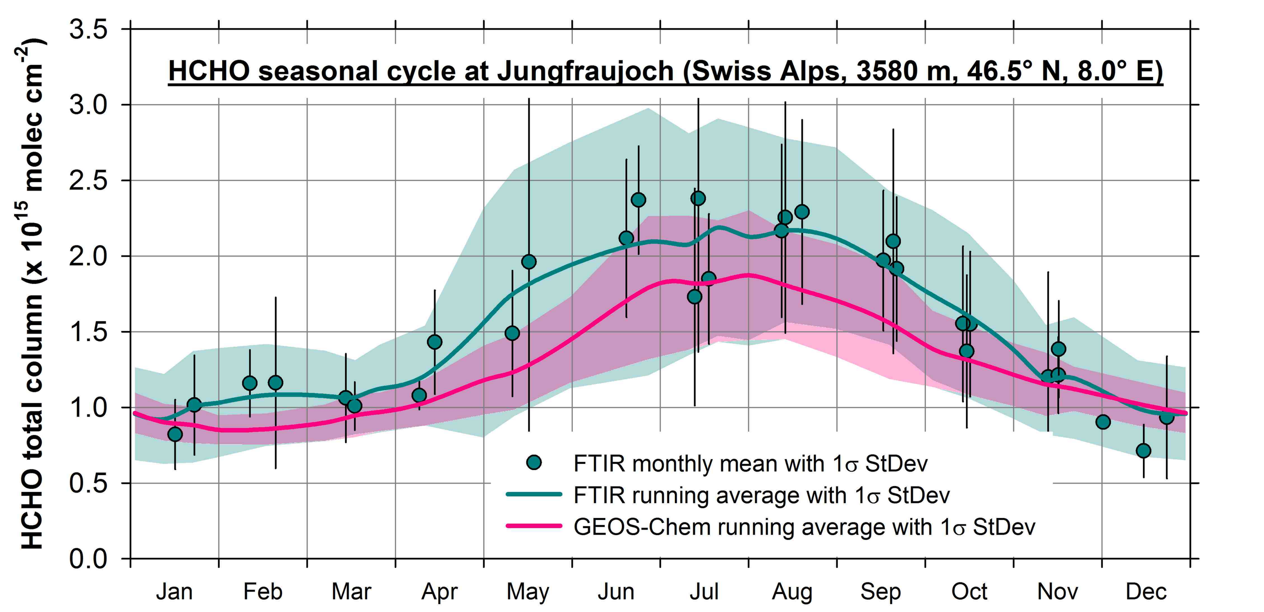


FIGURE 5 - Seasonal variation of formaldehyde as measured at the Jungfraujoch station (in green) and modeled by GEOS-Chem (v9-1-3; in red) over the mid-2010 - 2012 time period. We observe an underestimation of the summertime amount of formaldehyde that we hypothesize to be due to large uncertainties remaining in the emissions of HCHO precursors implemented by the model. An optimized retrieval strategy for HCHO from ground-based FTIR solar spectra has been developed and validated at Jungfraujoch. This strategy is implemented in an ongoing work which aims at exploiting the multi-decadal observational database available at Jungfraujoch (back to 1988 for HCHO) in order to investigate the interannual variability of formaldehyde, produce long-term trends and characterize its diurnal cycle in the remote atmosphere. Ground-based HCHO measurements are also increasingly required to validate satellite observations. For more details, see Franco et al., Atmos. Meas. Tech., 8, 1733-1756, 2015 (http://hdl.handle.net/2268/174025).

CURRENT LIST OF AVAILABLE TARGET GASES (JUNGFRAUJOCH)

Numerous atmospheric species have exploitable spectral signatures in the infrared region routinely recorded by the NDACC-affiliated ground-based FTIR instruments (see map). First priority species include O_3, HNO_3, HCl, HF, CO, N_2O, CH_4, HCN, C_2H_6 and $ClONO_2$. Total and partial column time series of all these species are available in hdf and/or NASA-Ames format from the NDACC database (http://www.ndacc.org).

Altogether, more than 30 molecules are now routinely retrieved from the Jungfraujoch spectra:

-- major greenhouse gases: H_2O, CO_2, CH_4 and N_2O
-- ozone (in the troposphere and stratosphere)
-- halogenated compounds: CCl_3F (CFC-11), CCl_2F_2 (CFC-12), $CHClF_2$ (HCFC-22), CH_3CClF_2 (HCFC-142b), CCl_4, CF_4, SF_6, HCl, $ClONO_2$, HF and COF_2
-- nitrogen compounds: N_2, N_2O, NO, NO_2, HNO_3, $ClONO_2$, NH_3
-- organic compounds: CO, C_2H_2, C_2H_4, C_2H_6, CH_3OH, HCN, formaldehyde, formic acid, OCS
-- many isotopologues of H_2O, CH_4, CO, O_3…

Currently under development: C_3H_8, PAN, CH_3Cl…

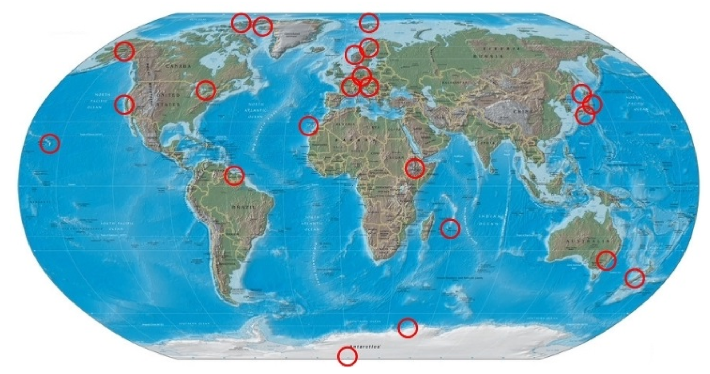


FIGURE 1 - NDACC FTIR sites location.

NETWORK & SITE, INSTRUMENTATION, OBSERVATIONAL DATABASE AND TOOLS

-- Very high resolution (up to 0.003 cm^{-1}) infrared solar spectra are recorded year-round, at the high-altitude International Scientific Station of the Jungfraujoch (Swiss Alps, 46.5°N, 8.0°E, 3580m a.s.l.). Clear-sky conditions are mandatory.

-- Fourier Transform InfraRed (FTIR) monitoring activities are conducted at that site within the framework of the Network for the Detection of Atmospheric Composition Change (NDACC, see http://www.ndacc.org). See **Figure 1** for a map of the current official NDACC sites.

-- Our FTIR instruments are equipped with cooled HgCdTe and InSb detectors, allowing covering the 650 to 4500 cm^{-1} region of the electromagnetic spectrum. A set of optical filters (color-coded in **Figure 2**) are used to maximize the signal-to-noise ratios.

-- The retrievals are essentially performed with the SFIT-2 algorithm (v3.91) which is based on the semi-empirical implementation of the Optimal Estimation Method of Rodgers [JGR, 95, 1990], allowing in most cases to retrieve some information on the vertical distribution of the target species.

-- Multidecadal FTIR time series are available from the Jungfraujoch (longest FTIR data sets worldwide), with earlier measurements in 1984.

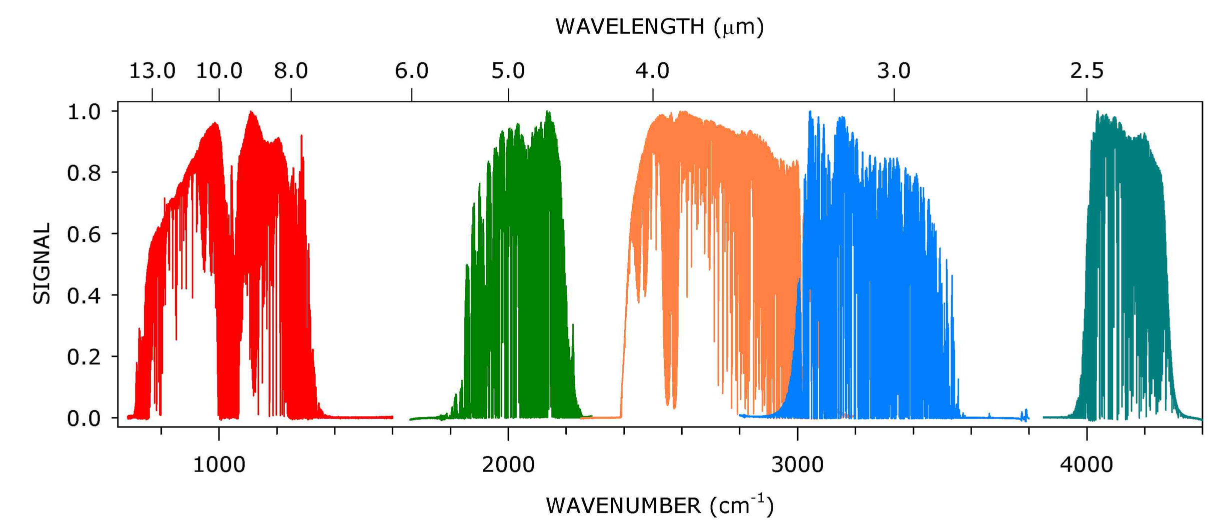


FIGURE 2 - Infrared spectral ranges routinely recorded at the Jungfraujoch station.

THE PyGChem PROJECT
(Benoît Bovy et al.)

PyGChem is a high-level, user-friendly Python interface under development for the GEOS-Chem model. Several tools and notebooks (data visualization & treatment) are available, they can be downloaded from the following dedicated site:

https://github.com/benbovy/PyGChem

ACKNOWLEDGMENTS

The University of Liège contribution to the present work has primarily been supported by the PRODEX and SSD programs (ACROSAT and AGACC-II projects, respectively) funded by the Belgian Federal Science Policy Office (BELSPO), Brussels. Laboratory developments and mission expenses were funded by F.R.S. – FNRS and the Fédération Wallonie-Bruxelles, respectively. We thank the International Foundation High Altitude Research Stations Jungfraujoch and Gornergrat (HFSJG, Bern) for supporting the facilities needed to perform the observations. E. Mahieu is Research Associate with the F.R.S. – FNRS. We further acknowledge the vital contribution from all colleagues in performing the FTIR observations used here.

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Poster presented at the 7th International GEOS-Chem Meeting, Harvard University, May 4-7, 2015