Overview of the recent results derived from the Jungfraujoch observational database

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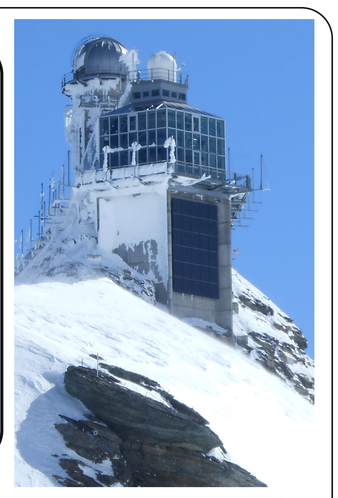


RECENT INCREASE OF ETHANE ABOVE NORTH AMERICA
(Franco et al., JQSRT, 160, 36-49, 2015)

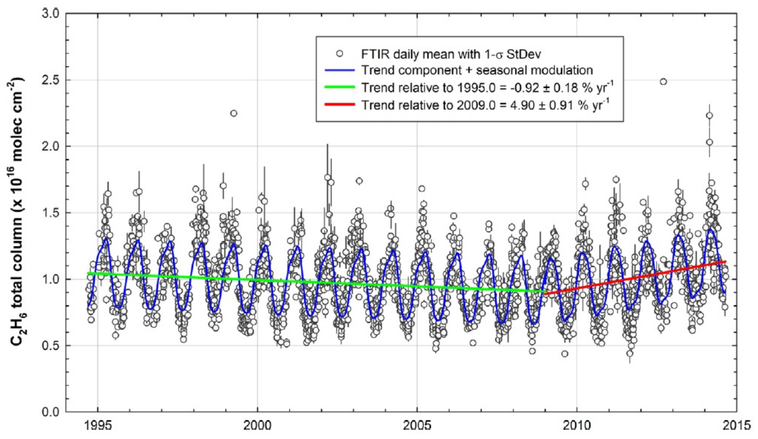


FIGURE 1 - Trends of ethane for Jungfraujoch, as deduced from long-term FTIR monitoring activities performed within the framework of the NDACC network. Consistent trends are observed at other sites (e.g. Boulder, CO, Toronto, ON) and confirmed by ACE-FTS occultation measurements above North America. 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.

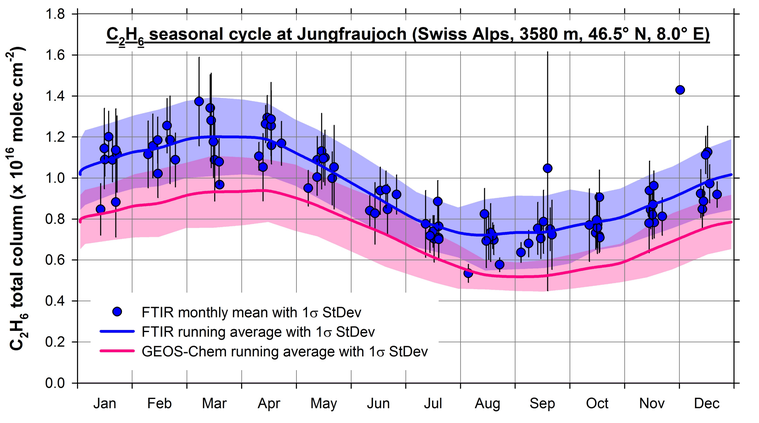


FIGURE 2 - Seasonal variation of ethane as measured at the Jungfraujoch station (in blue) and modeled by GEOS-Chem (v9-2; in red) for the days of observations over the mid-2005 - mid-2013 time period. We have taken into account the vertical resolution and specific sensitivity of the FTIR retrievals before 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 since they have been evaluated to 6%, with the major contribution originating from the uncertainty affecting the C_2H_6 spectroscopy, including the conversion to pseudolines parameters.

INVESTIGATING THE CAUSES FOR THE METHANE RISE AFTER 2005
(Bader et al., in preparation)

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.

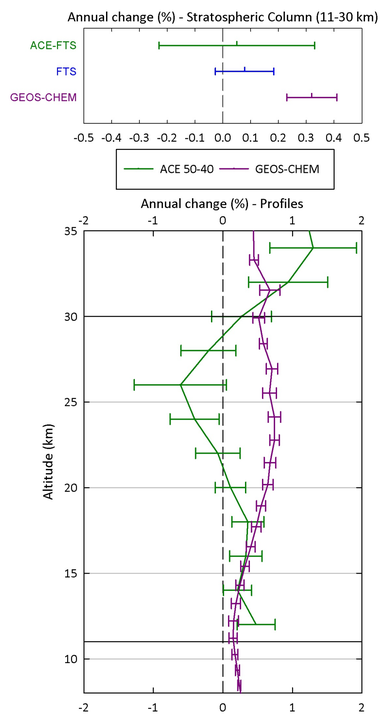


FIGURE 4 - Daily mean total column time series for the FTIR measurements (dark blue) and the GEOS-Chem simulation (in grey) are reproduced on the main frame, for the 961 available coincidences over the 2005-2013 (May) time period. Trends computed with a bootstrap resampling tool (Gardiner et al., *Atmos. 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 %.

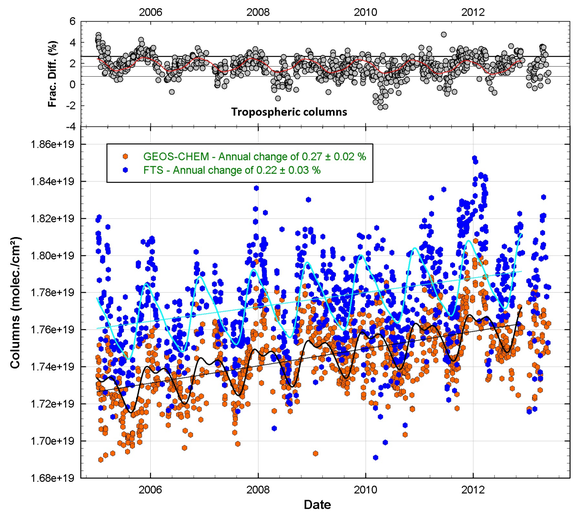


FIGURE 3 - A vertical bias between FTIR measurements and the GEOS-Chem simulation has been identified. It stands out that the annual changes of methane in the troposphere (3.58 - 11.7 km) computed from our measurements and GEOS-Chem (v9-2) simulation are in agreement, contrarily to the changes in the 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.

RETRIEVAL OF FORMALDEHYDE FROM AN UNPOLLUTED SITE
(Franco et al., AMT, 8, 1733-1756, 2015)

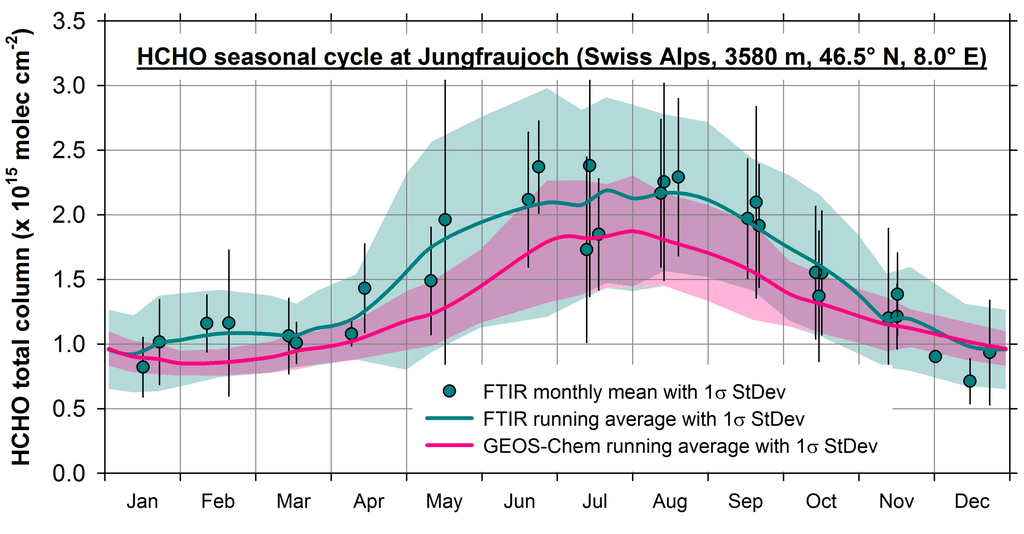


FIGURE 5 - Seasonal variation of formaldehyde as measured at the Jungfraujoch station (in green) and modeled by GEOS-Chem (v9-1-3; in red) over the mid-2010 - 2012 time period. We observe an underestimation of the summertime amount of formaldehyde that we hypothesize to be due to large uncertainties 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.

ATMOSPHERIC CIRCULATION CHANGES AND THEIR IMPACT ON HCl
(Mahieu et al., Nature, 515, 104-107, 2014)

As the most abundant reservoir of chlorine in the stratosphere, hydrogen chloride is an excellent indicator of the success of the Montreal Protocol on substances that deplete ozone. Time series of HCl monitored at NDACC FTIR stations are therefore scrutinized to characterize the evolution of the HCl loading in the Earth's atmosphere, and successive studies have reported about a stabilization of HCl in the atmosphere around the mid-1990s and of its subsequent decrease (see frame a of Figure 6). However, a recent and unexpected upturn in HCl has been detected, stimulating a study involving NDACC stations, a merged satellite data set (GOZCARDS, combining data from HALOE, ACE-FTS and Aura/MLS) and two chemistry transport models (KASIMA and SLIMCAT) to characterize this recent upturn and identify its cause.

Atmospheric circulation changes and variability have been identified as responsible for this unexpected feature. It was further possible to determine that a slowing of the Northern Hemisphere atmospheric circulation which occurred over a few year after 2005-2006 caused the recent rise in HCl. In the Southern Hemisphere, a rather constant speedup was diagnosed (see Figure 7).

The main conclusion of this study is that unidentified or unreported emissions of chlorinated source gases are not responsible for the recent rise in HCl in the Northern Hemisphere stratosphere. Hence, the Montreal Protocol is well on track and will lead to the reduction of the chlorine loading in the stratosphere, allowing ozone recovery in the next decades.

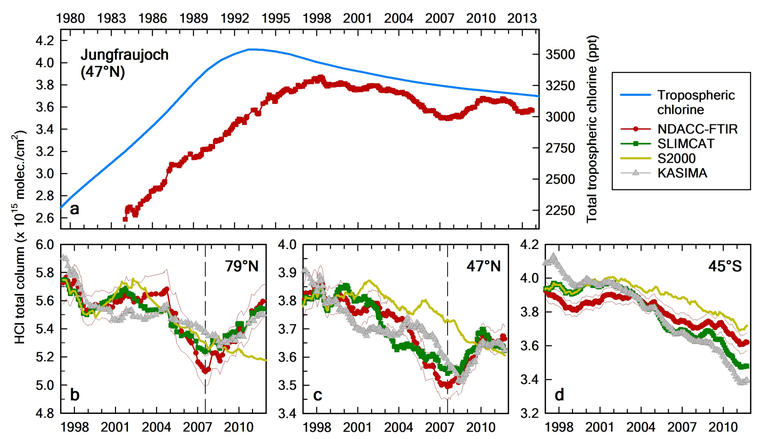
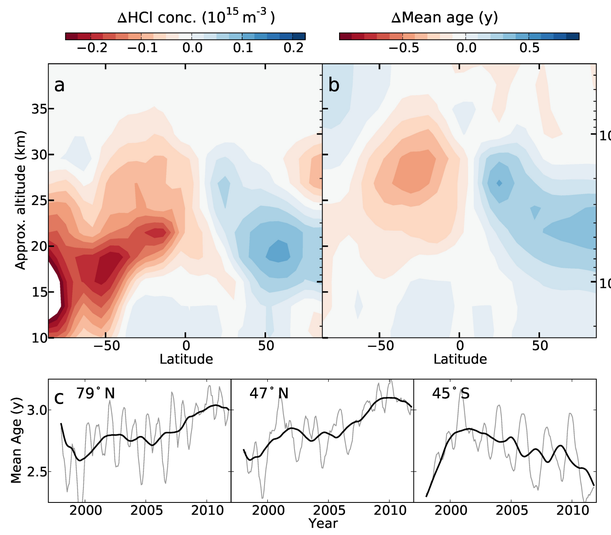


FIGURE 6 - The ground-based FTIR data sets (in red) consistently indicate that, after a period of decrease, the HCl started to increase again from July 2007 onwards in the Northern Hemisphere (see frame b and c for Ny Alesund and Jungfraujoch). Conversely, a continuous decrease was observed for the Southern Hemisphere (see frame d for Lauder). The SLIMCAT and KASIMA model simulations (in green and grey, respectively), using ERA-Interim meteorology and surface source gas mixing ratios from the WMO A1 scenario, were able to reproduce the specific situations prevailing in both hemispheres. In contrast, a dedicated SLIMCAT run (denoted S2000, light green) using 6-hourly winds of 2000 from 2000 onwards does not produce the recent HCl upturn. Comparison with sateliite data confimrs these findings (updated and adapted from Mahieu et al., nature13857, 2014).

FIGURE 7 - Spatial distribution of the HCl concentration and age-of-air changes. Mean differences of the HCl concentration (a) and age-of-air (b) between 2010/2011 and 2005/2006, as a function of altitude and latitude, derived from the standard SLIMCAT simulation (confirmed by KASIMA, not shown here). There is a clear asymmetry between both hemispheres, with correlated patterns between age-of-air and HCl, indicating that the HCl changes over that period are consistent with slower/faster circulation in the NH/SH. c. Running averages of the mean age-of-air at 50 hPa (thick/thin curve, integration length of 36/6 months), at the same sites as Fig. 6 (time series at 79°N and 45°S have been shifted vertically by -0.75 yr). The slower circulation in the NH lower stratosphere after 2005-2006 diagnosed here allowed a larger relative conversion of the chlorinated source gases into HCl, resulting in the HCl upturn (Mahieu et al., nature13857, 2014).



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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. We further acknowledge funding by the EU 7th Framework Programme project NORS (contract 284421) and MeteoSwiss (GAW-CH). 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 are very grateful to all the colleagues who performed the FTIR observations used here.

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Poster presented at the 2015 NDACC-IRWG meeting, University of Toronto, June 8-12, 2015