Name of research institute or organization:

Institut d'Astrophysique et de Géophysique, Université de Liège

Title of project:

High resolution, solar infrared Fourier Transform spectrometry. Application to the study of the Earth atmosphere

Part of this programme:

NDACC (Network for the Detection of Atmospheric Composition Change)
GAW (Global Atmosphere Watch)

Project leader and team:

Christian Servais (project leader), Benoît Bovy, Olivier Flock, Bernard Lejeune, Emmanuel Mahieu, Ginette Roland (em.), Vincent Van De Weerdt, Diane Zander

Project description:

The team's objectives are essentially twofold: (i) improve the performance of the instrumentation and perform the observations, (ii) analyse the spectra in order to produce high-level geophysical parameters and valorize them.

In 2016, observations have been performed on site or recorded through a remote-control web interface. Altogether, more than 2000 high resolution infrared solar spectra have been collected on 99 days. Our multidecadal observational program had to stop early December 2016. Indeed, the HFSJG Direction has initiated the renovation of the level 1 of the Sphinx observatory, involving dismounting a heavy part (a He-cooled unit) of the Fourier Transform InfraRed (FTIR) spectrometer, with possible impact on its intrumental profile. Resuming the observations will only be possible after a careful and thorough realignment of the instrument and instrumental profile check, in agreement with the NDACC-network best practices.

Greenhouse gases	H_2O, CO_2, CH_4, N_2O, CF_4, SF_6	Support to the Kyoto Protocol
Ozone-related	O_3, NO, NO_2, HNO_3, $ClONO_2$, HCl, HF, COF_2, CFC-11, CFC-12, HCFC-22, HCFC-142b, CCl_4, CH_3Cl	Support to the Montreal Protocol
Air quality	CO, CH_3OH, C_2H_6, C_2H_2, C_2H_4, HCN, HCHO, HCOOH, NH_3	Support to the EU-Copernicus programme
Other	OCS, N_2, various isotopologues	

Table 1. List of atmospheric species currently retrieved from the Jungfraujoch observational database.

Analysis of our spectra allows us to determine the abundance of an increasing number of constituents of the Earth atmosphere (currently more than 30; see Table 1), playing a role in ozone depletion, climate change or affecting air quality. Numerous target species are therefore relevant to the Montreal Protocol on substances that deplete stratospheric ozone (e.g. CFCs, HCFCs, HCl) and/or to the Kyoto Protocol on greenhouse gases emissions (e.g. CO_2, CH_4, N_2O). We briefly present hereafter two recent results derived from the exploitation of our database. For a complete view of the team's scientific output in 2016, the reader is invited to consult the literature (see the "Refereed journal articles" section).

First retrievals of HCFC-142b (CH_3CClF_2) from ground-based FTIR spectra

Hydrofluorocarbons (HCFCs) are the first substitutes to the long-lived manmade chlorofluorocarbons (CFCs). Following the progressive – and now complete – ban of the CFCs by the Montreal Protocol, emissions of the HCFCs have been on the rise. If the HCFC-

22 emissions remain the largest in this family with 366 Gg/yr at the present time, annual emissions of HCFC-142b still amount to ~30 Gg in 2012, after a maximum of ~40 Gg/yr in the late 2000s, justifying the monitoring of these ozone-depleting substances.

We have developed an approach for the retrieval of HCFC-142b from ground-based FTIR spectra, using the ν_7 band Q branch near 11µm. This was a challenging task since the absorption of HCFC-142b remains weak, below 1% at current abundances. We evaluated that HCFC-142b individual total columns are affected by random and systematic uncertainties of 19 and 9%, respectively.

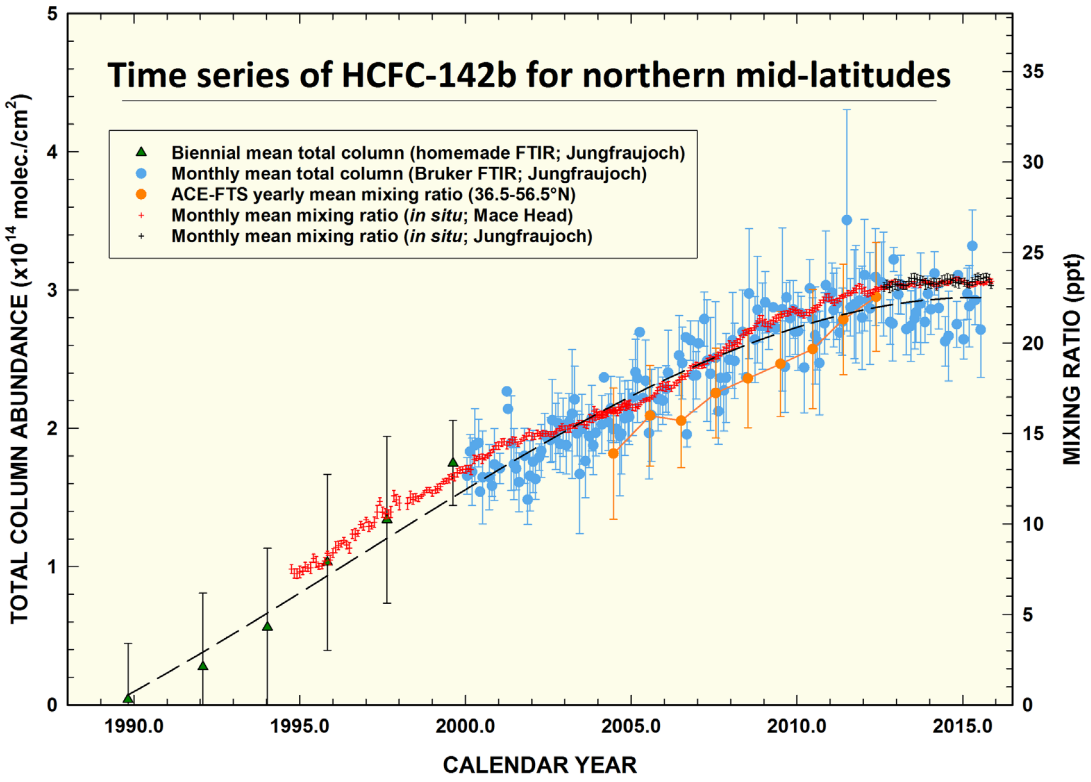

Figure 1. Time series of HCFC-142b for the northern mid-latitudes, as derived from infrared remote-sensing spectra and in situ surface measurements by the AGAGE network (see legend for their identification). The vertical axe provides the correspondence between the total columns and the surface mixing ratios (Mahieu et al., 2016).

The monthly mean FTIR total column time series derived from 2000 onwards is reproduced in Figure 1 as blue circles. Our results are compared with *in situ* surface measurements performed at Mace Head, Ireland (red crosses) and at Jungfraujoch by Empa (black crosses) as well as with satellite occultation measurements by the ACE-FTS instrument (Atmospheric Chemistry Experiment – Fourier Transform Spectrometer; orange circles). All data sets consistently show mean annual increases of HCFC-142b at rates of 0.8-1.0 ppt/yr for the 2000-2010 time period. Thereafter, a significant slowing down is observed, with annual increases in the 0.1-04 ppt/yr range over 2010-2014. More information is available in a recent dedicated publication (Mahieu et al., 2016).

Revisiting the OCS (carbonyl sulfide) retrieval strategy

Carbonyl sulfide (OCS) is a major source of sulfur to the stratosphere, sustaining the Junge (aerosol) layer and therefore playing a role both in climate and in stratospheric ozone chemistry. Investigating atmospheric OCS is also justified because of its uptake by vegetation. Indeed, since it is – in contrast to CO_2 – not emitted by respiration, OCS has the potential to provide useful constraints on the photosynthesis flux, while when studying CO_2,

only the net signal can be derived, without clear indication of the respective contribution of photosynthesis and respiration (e.g., Wang et al., 2016).

Lejeune et al. (2016) have determined an optimized approach for the retrieval of OCS, after a careful, systematic and objective evaluation of a significant subset (117) of all OCS lines belonging to the ν_3 fundamental band. The updated strategy provides a better representation of the seasonal modulation of OCS and limits the impact of the H_2O and CO_2 interferences. With a typical DOFS (Degree of Freedom for Signal) of 2.8, robust information is available for both the tropospheric and stratospheric abundances of OCS.

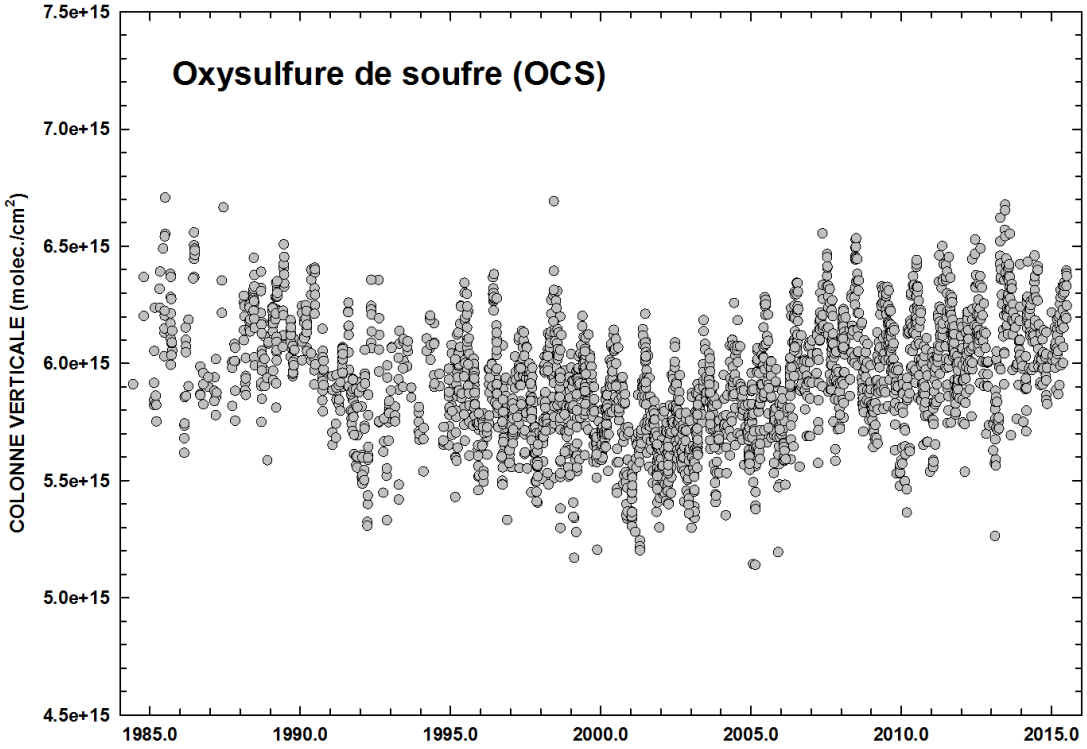

Figure 2. FTIR time series of daily mean OCS total columns over the last 30 years as derived from the Jungfraujoch observational database (updated from Lejeune et al., 2016).

Figure 2 reproduces the time series of OCS daily mean total columns available from the Jungfraujoch station. It is the longest available worldwide, with data now covering more than 3 decades. This data set indicates that the abundance of OCS has been stable since 2009. Beforehand, a significant decrease has been characterized between 1984 and 2002 (-2.7×10^{13} molec./cm^2 per year, or -0.5 %/yr when considering the mean OCS total column over 1984-2015), followed by an increase until 2008 ($+6.9\times10^{13}$ molec./cm^2 per year, or +1.2 %/yr). Lejeune et al. (2016) have shown that the tropospheric columns have driven these changes, the OCS loading remaining essentially constant in the stratosphere over 1984-2015. The causes of the observed changes remain a matter of debate, but the evolution of the emissions associated with the rayon production (releasing CS_2) and coal combustion are likely at play. Model simulations will be needed to confirm this hypothesis.

Key words:
Earth atmosphere, climate change, greenhouse gases, ozone layer, air quality, long-term monitoring, infrared spectroscopy, atmospheric circulation

Internet data bases:

General website: http://girpas.astro.ulg.ac.be
Consolidated geophysical data are available from NDACC:
ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/jungfrau/hdf/ftir/

Collaborating partners/networks:

Main collaborations: BIRA-IASB (Institut d'Aéronomie Spatiale de Belgique) / NDACC (Network for the Detection of Atmospheric Composition Change; http://www.ndacc.org) / GAW-CH / ACE science team / NASA JPL / Empa / University of Leeds / IMK (Forschungszentrum Karlsruhe) / satellite experiments: IASI, OMI, ENVISAT / …

Scientific publications and public outreach 2016:

The complete list of GIRPAS publications can be found at
http://labos.ulg.ac.be/girpas/en/publications

Refereed journal articles and their internet access

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