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AGACC-II:

**Advanced exploitation of Ground-based measurements for Atmospheric Chemistry
and Climate applications - II**

SCIENCE FOR A SUSTAINABLE DEVELOPMENT

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ACRONYMS, ABBREVIATIONS AND UNITS

AERONET	AERosol RObotic NETwork
AGACC	Advanced exploitation of Ground-based measurements for Atmospheric Chemistry and Climate applications
AGAGE	Advanced Global Atmospheric Gases Experiment
AOD	Aerosol Optical Depth
BIRA-IASB	Belgisch Instituut voor Ruimte-Aeronomie – Institut d’Aéronomie Spatiale de Belgique
CFC	ChloroFluoroCarbon
[CHIMERE	Name of a multi-scale chemistry-transport model for air quality forecasting and simulation.]
[CIMEL	Commercial provider of sunphotometers]
CTM	Chemistry-Transport Model
EGU	European Geosciences Union
FIR	Far-InfraRed
FTIR	Fourier Transform InfraRed
FTS	Fourier Transform Spectrometer
FUNDP	Facultés Universitaires Notre Dame de la Paix
GCOS	Global Climate Observing System
GEISA	Gestion et Etude des Informations Spectroscopiques Atmosphériques (Management and Study of Atmospheric Spectroscopic Information)
GHG	GreenHouse Gas
GWP	Global-Warming Potential
HITRAN	High resolution TRANsmision database
IASI	Infrared Atmospheric Sounding Interferometer
ICOS	Integrated Carbon Observing System
IR	InfraRed
JQSRT	Journal of Quantitative Spectroscopy and Radiative Transfer
KMI-IRM	Koninklijk Meteorologisch Instituut – Institut Royal Météorologique
LACy	Laboratoire de l’Atmosphère et des Cyclones
LSCE	Laboratoire des Sciences du Climat et de l’Environnement
mK	milliKaiser (0.001 cm^{-1})
MAXDOAS	MultiAXis Differential Optical Absorption Spectroscopy
MIR	Mid-InfraRed
NDACC	Network for the Detection of Atmospheric Composition Change
NIR	Near-InfraRed
PAN	PeroxyAcetyl Nitrates
PBL	Planetary Boundary Layer
PI	Principal Investigator
[PICARRO	Commercial provider of greenhouse gas analyzers]
pptv	Parts Per Trillion by Volume
SSA	Single Scattering Albedo
STCE	Solar-Terrestrial Centre of Excellence
TCCON	Total Carbon Column Observing Network
TDL	Tunable Diode Laser
ULB	Université Libre de Bruxelles
ULg	Université de Liège
UNEP	United Nations Environment Programme
UV	UltraViolet
VOC	Volatile Organic Compound
WMO	World Meteorological Organization
WOUDC	World Ozone and Ultraviolet Radiation Data Centre
WP	Work Package

1. EXECUTIVE SUMMARY

All AGACC partners have already achieved some interesting results, at least intermediary.

Greenhouse gases

A dedicated spectrometer for making observations of greenhouse gases in the near-infrared spectral range, in the frame of the Total Carbon Column Observing Network (TCCON), has been operational at Ile de La Réunion since September 2011, and the first series of CH₄ and CO₂ data seem to have the required quality.

Historical measurements of greenhouse gases made at the Jungfraujoch with a grating spectrometer have been analysed and combined with the more recent timeseries from the later Fourier transform spectrometers. Once this work is completely finished, the resulting timeseries will cover the period from 1977 onwards, allowing to nicely extend the Network for the Detection of Atmospheric Composition Change (NDACC) mid-infrared data sets.

At St Denis at Ile de La Reunion, a special campaign has been conducted to evaluate the consistency between the NIR and MIR measurements of CH₄. First results of that evaluation are expected by April 2012.

New strategies for the retrieval of CCl₄ and CF₄ from Jungfraujoch FTIR spectra have been developed and implemented. The resulting timeseries (CCl₄ from 1999 until now, and CF₄ from 1990 until now) have been investigated. Two corresponding peer-reviewed publications are underway.

New volatile organic species

At Ile de La Réunion, we developed or improved retrieval strategies for formaldehyde, methanol, and ethane, and the resulting time series since 2004 have been compared to model simulations (IMAGESv5), showing that the models in general underestimate the seasonal variation due to biomass burning emissions. Additional biomass burning species (formic acid, hydrogen cyanide and acetylene) have been investigated at the same time, although they were not part of the AGACC-II objectives.

At Jungfraujoch, work has been done about the retrieval of methylchloride. Despite the use of improved spectroscopic data for ethane, an interfering species in the fitted microwindow, it is still difficult to obtain satisfactory results. This study will nevertheless be continued.

New and improved laboratory spectroscopic data

Dedicated laboratory spectra have been recorded for investigating the 10 µm region of ¹²C₂H₄, the ν₅ and ν₇ bands (3.3 µm region) of ¹²C₂H₆ and the ν₁ and ν₅ bands (3.6 µm region) of H₂ ¹²C¹⁶O. The analysis of the spectra is ongoing.

Once the final results for the spectroscopic parameters will become available, they will be used in the retrievals of the field observations at Jungfraujoch and Ile de La Réunion.

Aerosol investigations at Ukkel

A new cloud-screening algorithm has been developed and tested for deriving the Aerosol Optical Depth (AOD) at 340 nm from the Brewer measurements. Now, a very good agreement between simultaneous CIMEL sunphotometer and Brewer measurements is obtained.

A new ceilometer has been installed and operated in Ukkel to derive the aerosol backscatter ratio profiles and additional parameters (PBL, cloud information) .

CIMEL sunphotometer and Brewer measurements have been continued. Once we have a more complete set of aerosol observations (expected in 2012), we expect more interesting data and information about the aerosol and their radiative forcing at Ukkel.

As the project evolves and intermediary results become available, more exchanges between the partners and the various activities will become feasible.

2. INTRODUCTION

2.1 Context

The AGACC-II research contributes to international ground-based remote sensing networks and their expansion, in terms of capabilities and in terms of sites. It also contributes to global change research in general. It includes laboratory experiments in support of the remote sensing observations. It combines the expertise in the field of remote sensing and laboratory spectroscopy support of the 4 Belgian partners involved in the project, and that of the networks in which the work is embedded. Moreover the partners have strong collaborations with satellite teams and the atmospheric modeling community, for the interpretation of the observations.

The project also wants to start field observations in Africa, and therefore to initiate a collaboration with the University of Bujumbura in Burundi.

2.2 Objectives

1. The first major objective is to improve or expand the measurement capabilities for greenhouse gases (CO_2 , CH_4 and N_2O , as well as CF_4 and CCl_4) with FTIR spectrometers at Jungfraujoch and Ile de La Réunion, to make Ile de La Réunion a site affiliated to the Total Carbon Column Observing Network (TCCON), and to deliver long-term trends of the major greenhouse gases at the Jungfraujoch back to 1976.
2. Secondly, we aim at extending the measurement capabilities of ground-based FTIR spectrometry to various volatile organic compounds (CH_3Cl , CH_3OH , PAN, acetone) and to some CFC-substitutes, and consequently, using also the results for CCl_4 and CF_4 mentioned above, to establish representative CCl_y and CF_y budgets at the Jungfraujoch, including the contributions from the new AGACC-II halocarbons and of the standard FTIR products (CFC-11 and -12, HCFC-22). It is also planned to revise the time series for C_2H_4 , C_2H_6 , H_2CO based on better laboratory data, at Jungfraujoch and Ile de La Réunion.
3. A third objective is to provide improved laboratory spectroscopic data in support of the above objectives 1 and 2.
4. Fourth, we aim at advancing our understanding of aerosol characteristics above Ukkel and to estimate the aerosol direct radiative forcing above Ukkel. Therefore the underlying objectives are
 - to retrieve more information regarding aerosol properties from remote sensing measurements with Brewer and MAXDOAS spectrometers;
 - to deploy a lidar ceilometer at Ukkel;
 - to combine the information from all instruments including those from the CIMEL sunphotometer, for a more comprehensive evaluation of the aerosol properties at Ukkel;
 - to derive information about the aerosol sources by modeling with CHIMERE and backtrajectory studies;
5. The fifth objective is to implement MAXDOAS measurements in Bujumbura (Burundi, Africa) for the measurement of aerosol and ozone precursors (NO_2 , glyoxal, H_2CO ,...) and to study the export of African emissions to the Indian Ocean (Ile de La Réunion).
6. And finally, we want to make sure that the results of the project are disseminated appropriately and that they are integrated in national and international environmental assessments in support of policy makers.

2.3 Expected outcomes

The new data that we plan to produce in AGACC-II will be very useful for global and regional chemistry-climate models, as they provide means to verify the models that are used for understanding atmospheric processes and biogeochemical cycles, and for forecasting the future state of the atmosphere and climate. Moreover, the laboratory and the field data will be delivered to international databases like the HITRAN and GEISA databases, and the NDACC and TCCON databases, respectively. They will also be integrated in regional and global assessment reports. Some of the results will appeal directly to policy makers: long-term trends of greenhouse gases, aerosol direct radiative forcing above Ukkel, organic chlorine and fluorine budgets, ...But on the longer term, all the AGACC-II results will be relevant to environmental policies, because monitoring of atmospheric composition changes is a fundamental ingredient of any environmental assessment in support of policy makers.

The proposed research is also embedded in the international research programmes to which the partner institutes contribute and it will involve interactions with the international research community. It will strengthen the role of Belgium in NDACC. Moreover, it will enable Belgian researchers to join TCCON, another important international observation network cooperating with NDACC, which will probably be integrated in the near-future in ICOS, the Integrated Carbon Observing System.

3. SCIENTIFIC METHODOLOGY

Overall methodology

In AGACC-II, several ground-based remote sensing observation techniques are further developed, improved and exploited for the study of the Earth atmosphere composition and the link with climate. These techniques are Brewer, MAXDOAS and Fourier transform infrared (FTIR) spectrometry, as well as sunphotometry. An aerosol lidar (ceilometer) has been installed at Ukkel in 2011 and is now operational as well. Most of the instruments exploited in the project are integrated in global observation networks, like the Network for the Detection of Atmospheric Composition Change (NDACC), and AERONET. The observation sites included in the project are Ukkel (50.5°N, 4°E), Jungfraujoch (46.5°N, 8°E), and Ile de La Réunion (21°S, 55°E), as was the case previously; we hope to start working also in Bujumbura (3°S, 29°E) in Africa where we plan to install a MAXDOAS spectrometer in 2012.

The project also includes dedicated laboratory experiments to support the field observations, and modeling exercises (via collaborations) for an enhanced interpretation of the measurements.

Dedicated tasks

WP1 - Exploitation of FTIR spectra to derive useful geophysical information for a suite of GHGs (CO₂, CH₄, N₂O, CF₄, CCl₄)

Task 1.1. Produce time series for the major GHGs CO₂, CH₄ and N₂O

Task 1.1.1 Analyse historical grating spectra at the Jungfraujoch between 1976 and 1989, merge them with more recent data and derive long-term trends of the major greenhouse gases CO₂, CH₄ and N₂O above the Jungfraujoch.

Historical solar spectra recorded at the Jungfraujoch with a high-resolution grating spectrometer have been re-analyzed to derive total columns of a series of atmospheric gases. This instrument was used in the Sixties and Seventies to record two solar spectrum atlases extending from the near-ultraviolet to the near-infrared (from ~300 to ~1200 nm). From 1977 to 1989, it was also regularly and intentionally used to record narrow spectral intervals in the mid-infrared, encompassing absorption lines of gases of atmospheric interest, e.g. CH₄, HF, HCl, H₂O, N₂O, NO₂, C₂H₆, O₃ and CO. More than ten thousand spectra were recorded during this period.

In the 1970s – 1980s, the plotted grating spectra were analyzed by manually measuring the equivalent width of the target gas lines with a high-precision Coradi planimeter. They were never analyzed with fitting programs, which did not exist at that time. An effort has been undertaken to cautiously and consistently re-analyze and valorize these early observations, with modern spectral fitting tools, in particular SFIT-2.

The total columns derived from the grating spectra have been combined –after careful intercalibration using common observation days– with the FTIR columns derived at the Jungfraujoch since the mid-1980s, in order to produce consistent datasets and to derive the temporal evolution of various target gases for the period 1977-2011. Although this effort is ongoing, preliminary results obtained for CH₄ and N₂O can already be shown → see chapter 4.

Details on the strategies used to retrieve CH₄ total columns from the grating spectra can be found in Bader et al. (2011) and in Bader (2011).

As to nitrous oxide investigations, several N₂O lines located in the 2481 – 2483 and 2537 – 2540 cm⁻¹ spectral intervals were used (resolution between 6.9 and 10.3 mK, depending on the grating order and slit width). Inter-comparison between columns derived from coincident homemade FTS and 2482 cm⁻¹ grating observations, as well as between 2538 and 2482 cm⁻¹ grating spectra showed very good agreements, with ratios of 1.00 ± 0.01 and 1.00 ± 0.02, respectively. This gives strong confidence when it comes to combine the various subsets in order to derive long-term trends.

Task 1.1.2 Implement TCCON-compliant measurements at Ile de La Réunion for the measurement of CO₂, CH₄ and N₂O.

In September 2011, we started FTIR measurements using a new Bruker 125/HR spectrometer, solar tracker and meteorological station, compliant with the requirements of TCCON (Total Carbon Column Observing Network) in St. Denis at Ile de La Réunion. This new instrument performs measurements in the near- and mid-infrared region for the TCCON and NDACC networks, respectively. Since then, near-infrared spectra have been analyzed according to the TCCON guidelines, and the first results for

the total column abundances of CO₂ and CH₄ have been obtained (see Chapter 4). Our observatory at La Réunion is now the fourth operational TCCON station in the Southern Hemisphere. We are still working to obtain the official TCCON quality label – which requires in principle an aircraft flight with in-situ measurements of CH₄ and CO₂ above the instrument site, to verify the calibration of our TCCON data. We are discussing with our French partner at La Réunion the organization of such a flight.

In parallel, we are equipping the site with an in-situ greenhouse gas monitoring instrument (PICARRO, as required in the context of ICOS) which will become operational in the course of 2012. This will also help us to verify the absolute calibration of our TCCON data.

Task 1.1.3 Investigate the consistency between measurements of CH₄ in the near infrared (NIR) (6000 cm⁻¹), the 4000-4400 cm⁻¹ region and the mid-infrared (MIR) (2650 cm⁻¹), and verify the precision obtained in the different spectral ranges

From September to December 2011, our new Bruker 125/HR and the 'old' Bruker 120/M (operating in the mid-infrared), which has since then been decommissioned, operated side-by-side at La Réunion. At present, the results obtained by the two instruments are being analyzed in the framework of the detailed comparison between both instruments and between the mid-infrared and the near-infrared retrievals for CH₄.

Task 1.2 Investigate additional GHGs: CCl₄ and CF₄

Carbon tetrachloride (CCl₄)

The most appropriate feature for the retrieval of CCl₄ from ground-based FTIR spectra is the strong ν_3 band near 12.7 μm . Thus far however, such retrievals have been hampered by a nearby CO₂ Q-branch affected by spectroscopic line-mixing. Neglecting this effect resulted in large systematic residuals (see frame B of **Figure 1**), with expected negative influence on the retrievals of CCl₄ and on its error budget.

We have therefore implemented in the SFIT-2 algorithm a routine of J.-M. Hartmann (LISA, IPSL, Paris) to account for line-mixing effects in the CO₂ Q branch at 792 cm⁻¹. Improvement in the residuals is obvious when comparing frame A and B of **Figure 1**.

A retrieval strategy further accounting for interferences by water vapor and ozone has been set up, with the fitted microwindow extending from 785 to 807 cm⁻¹. Careful error budget evaluations have indicated for a single fit a total random error of less than 7%, a total systematic error of less than 11%, the latter being essentially influenced by the uncertainty on the CCl₄ spectroscopic line parameters.

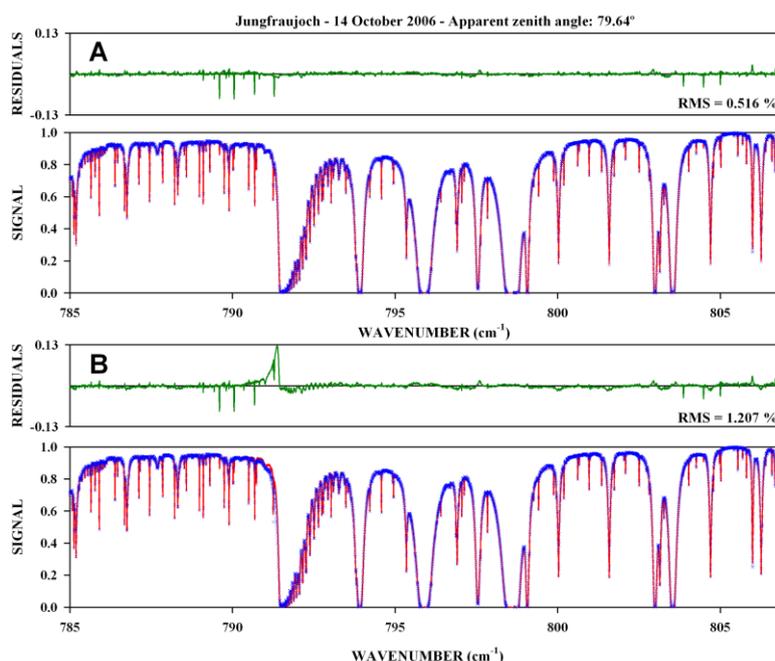


Figure 1. Sample fits to a spectrum recorded on October 14, 2006 at the Jungfraujoch station. The observed and fitted spectra are reproduced in red and blue, respectively. Frame A displays in green the observed minus calculated residuals when accounting for the line mixing in the CO₂ Q branch. This corresponds to a significant improvement when compared to the residuals which were obtained when neglecting this spectroscopic effect (see frame B).

Carbon tetrafluoride (CF₄)

For the first time, CF₄ has been retrieved from ground-based FTIR spectra. A two-step retrieval scheme has been developed to determine CF₄ total columns, focusing on the strong ν_3 band around 1282 cm⁻¹. Numerous interferences have to be accounted for, among which: several isotopologues of H₂O, N₂O, ¹³CO₂, ClONO₂, HNO₃ and H₂O₂. In particular, H₂¹⁶O and HDO interferences strongly reduce the fitting quality during wet days. Consequently, we had to discard the wettest observations from our data base.

WP 2 - Volatile Organic Compounds (VOC) and CFC-substitutes in the troposphere

Task 2.1 Develop retrieval strategies for methanol (CH₃OH) and methylchloride (CH₃Cl) at Jungfraujoch and La Réunion, and analyse the available time series. Model and satellite comparisons will be performed if possible.

Methyl chloride (CH₃Cl) at Jungfraujoch

Methyl chloride is one of the most abundant chlorine-bearing gas in the Earth's atmosphere and a significant contributor to the organic chlorine budget, with mean volume mixing ratio of 550 pptv. Several natural sources have been identified (e.g. dead leaves in tropical regions, biomass burning) while the largest sink is oxidation by OH. Although balanced, its atmospheric budget is still affected by significant uncertainties.

There are numerous but weak features of CH₃Cl near 3µm. Unfortunately, experimental spectra indicate that line parameters corresponding to many CH₃Cl features are missing in HITRAN, even in the most recent edition. This was also true for ethane until very recently. Fortunately, pseudoline parameters for ethane have been produced on the basis of Harrison et al. (2010) cross section measurements. Mahieu et al. (2011b) have shown that these new parameters allow to significantly improve the quality of the fits, with root-sum-square residuals nearly divided by three in the window we selected for retrieving CH₃Cl (between about 2966 and 2968 cm⁻¹).

Despite this improvement, the retrieval of CH₃Cl from ground-based FTIR spectra remains very challenging. A subset of Jungfraujoch spectra has been fitted to retrieve total columns of methyl chloride. First results were not in line with expectations: retrieved columns are on average about 50% larger than anticipated, furthermore, the strong seasonal signal characterizing our preliminary time series is out of phase (maximum columns are found in February, i.e. when surface concentrations derived from in situ samplings are maximum). Further investigations will be needed to determine what might be the causes of the present discrepancies.

Methanol (CH₃OH) and other VOC at Ile de La Réunion

The retrieval strategies for CH₃OH and C₂H₆ have been optimized for Reunion Island. The spectral micro-windows are 1029-1037 cm⁻¹ for methanol, and 2976.66-2976.95 cm⁻¹ and 2983.20-2983.55 cm⁻¹ for C₂H₆. The spectroscopic database HITRAN 2008 (Rothman et al., JQSRT, 2009) is used for methanol while for C₂H₆ we have used pseudo-lines constructed by G. Toon (private communication) based on the recent paper of Harrison et al. (2010). However, we expect that the work of the ULB spectroscopic team, when available, will improve again the retrieval results. Water vapor being a strong interfering gas at Reunion Island, preliminary retrievals of H₂O were made before the CH₃OH and C₂H₆ retrievals. For CH₃OH and C₂H₆, we also needed H₂¹⁸O and O₃ preliminary retrievals, respectively.

The strategy of HCHO was optimized and published in Vigouroux et al. (2009). Small changes have been made since then: the micro-windows for the preliminary retrievals of the interfering gas H₂O and HDO have been changed. The micro-window for HDO being now the 2855.65-2856.4 cm⁻¹, it has been removed from the previous set of HCHO micro-windows used in Vigouroux et al. (2009).

Task 2.2 Re-analysis of C₂H₄, C₂H₆ and H₂CO data at Jungfraujoch and La Réunion with improved spectroscopic data

Task 2.2.1 Provide improved laboratory spectroscopic data for ¹²C₂H₄, ¹²C₂H₆ and H₂¹²C¹⁶O

Remote sensing measurements are bound to the availability of reference spectroscopic information, and the final accuracy of the retrieved atmospheric parameters is obviously influenced by the quality of these reference data. Such reference information, measured in the laboratory and usually gathered in

databases such as HITRAN (Rothman et al, 2009), can only evolve slowly, adding new molecules and replacing existing parameters with better accuracy information. This situation is the combined consequence of the difficulty in accurately measuring spectroscopic parameters, and the intimate link between these activities and technological progress and financial support.

Within AGACC-II, in addition to answering to specific needs of the partners for reference spectroscopic information as they emerge, ULB has committed itself to try and improve line intensities for ethylene in the 10 μm spectral region, line positions and intensities for ethane in the 3.3 μm region, and line intensities for formaldehyde in the 3.6 μm region. The activities initiated and carried out by ULB in 2011 and results obtained are described hereafter. These activities are all ongoing.

Usually, the experimental work carried out at ULB is combined with theoretical support made available through collaborations. Indeed, theoretical models are in many cases essential to generate lists of line parameters (positions, intensities, widths...) useful for the analysis of atmospheric spectra. The theoretical models involved within AGACC-II describe the energy levels structure of molecules and transitions between them, to predict line positions and intensities. These predictions can be made, provided the values of the parameters involved in these models have been determined using line positions and intensities measured in the laboratory.

Task 2.2.1.a Provide improved laboratory spectroscopic data for $^{12}\text{C}_2\text{H}_4$

The aim of this task is to determine whether line intensities presently in HITRAN (Rothman et al, 2009) for the 10 μm spectral range of ethylene need improvements, and proceed if such is the case. This activity is carried out with theoretical support provided by two research teams, at the *Université de Reims Champagne Ardenne* (Prof. M. Rotger, Reims) and at the *Université de Bourgogne* (Dr V. Boudon, Dijon). The 10 μm region corresponds to the excitation of the ν_{10} , ν_7 , ν_4 and ν_{12} modes of vibration, whose fundamental bands are observed near 826, 949, 1026 and 1442 cm^{-1} , respectively.

We recorded two sets of absorption spectra of the 10 μm region, using a high resolution Fourier transform spectrometer Bruker IFS125HR available in our laboratory. The first set consisted of 6 spectra of ethylene contained in a 1.46-cm long cell, whose temperature was stabilized at 296 K, at pressures in the range 1.6 – 9.9 hPa. These spectra are aimed at providing line intensities for the ν_7 band, the strongest in that spectral range. We recently recorded a second set of 3 spectra at room temperature (293 K), with an absorption path length of 698 cm, and at pressures of 0.13, 0.38 and 0.67 hPa. These conditions are tailored to the observation of weak to very weak lines of the ν_{10} and ν_4 bands, and the measurement of their positions.

Task 2.2.1.b Provide improved laboratory spectroscopic data for $^{12}\text{C}_2\text{H}_6$

The 3.3 μm region of the spectrum of ethane is dominated by the ν_5 and ν_7 bands, respectively observed near 2896 and 2985 cm^{-1} , the latter exhibiting very strong Q branches involving lines blended even at Doppler-limited resolution (Pine & Lafferty, 1982). The reference spectroscopic information available for this spectral region is limited, mainly because of the absence of appropriate theoretical modeling. This situation results from the extreme complexity of the energy levels structure involved. Relying on a theoretical model developed by the group of C. di Lauro (Università di Napoli Federico II), the present contribution is an attempt to improve this situation.

We recorded 4 high-resolution spectra in the 3.3 μm region of ethane contained in either a 19.7-cm or a 5.15-cm long cell, at 296, 280 or 229 K and pressures from 0.2 to 1.4 hPa, using a Bruker IFS125HR Fourier transform spectrometer. Relying on line positions measured in the spectrum recorded at 229 K, together with those available from a spectrum recorded at 119 K (Pine & Lafferty, 1982), an analysis of the ν_7 band was attempted. Considering the more important interactions affecting the upper state of the ν_7 band, the theoretical model included 5 vibrational states.

Task 2.2.1.c Provide improved laboratory spectroscopic data for $\text{H}_2^{12}\text{C}^{16}\text{O}$

The 3.6 μm spectral region corresponds to the ν_1 and ν_5 bands, respectively located near 2782 and 2844 cm^{-1} , for which absolute line intensities have recently been reported (Perrin et al, 2009). Because formaldehyde polymerizes or degrades easily, the gas phase in the cell may contain various products which may bias the particle density of formaldehyde determined from measurements of the total pressure. In the present contribution, we aim at deriving infrared line intensities for formaldehyde using intensities of pure rotation lines, observed in the far infrared range, and the known electric dipole moment to determine its particle density. As a result, spectra in the infrared (IR) and far infrared (FIR) spectral regions had to be recorded simultaneously.

These recordings were carried out at ULB, in the FIR range using the Bruker IFS125HR Fourier transform spectrometer available and in the IR region using a tunable diode laser (TDL) spectrometer available in the group of Prof. M. Lepère (*Facultés Universitaires Notre Dame de la Paix, FUNDP, Namur*), installed at ULB. **Figure 2** shows the arrangement of the two spectrometers during the measurements. The formaldehyde sample (~1.5 % diluted in 10 Torr of N₂) was contained in a cross-shaped stainless-steel cell (shown in the inset in Fig. 3) located inside the evacuated FTS. The path lengths in the cross-shaped cell were 271 and 812 mm for the FIR and IR beams, respectively. A total of 5 experiments have been conducted at 296 K with 5 different samples of formaldehyde. Each experiment lasted about 8 hours, which was the time needed to record about 50 interferograms to yield one high-resolution FIR spectrum in the range 0–400 cm⁻¹ (the range useful for the present study of the pure rotation spectrum of H₂CO being 25–55 cm⁻¹). In the meantime, about 30 IR spectra were recorded around 2920 cm⁻¹ using the TDL spectrometer, each requiring about 1 minute to be obtained.

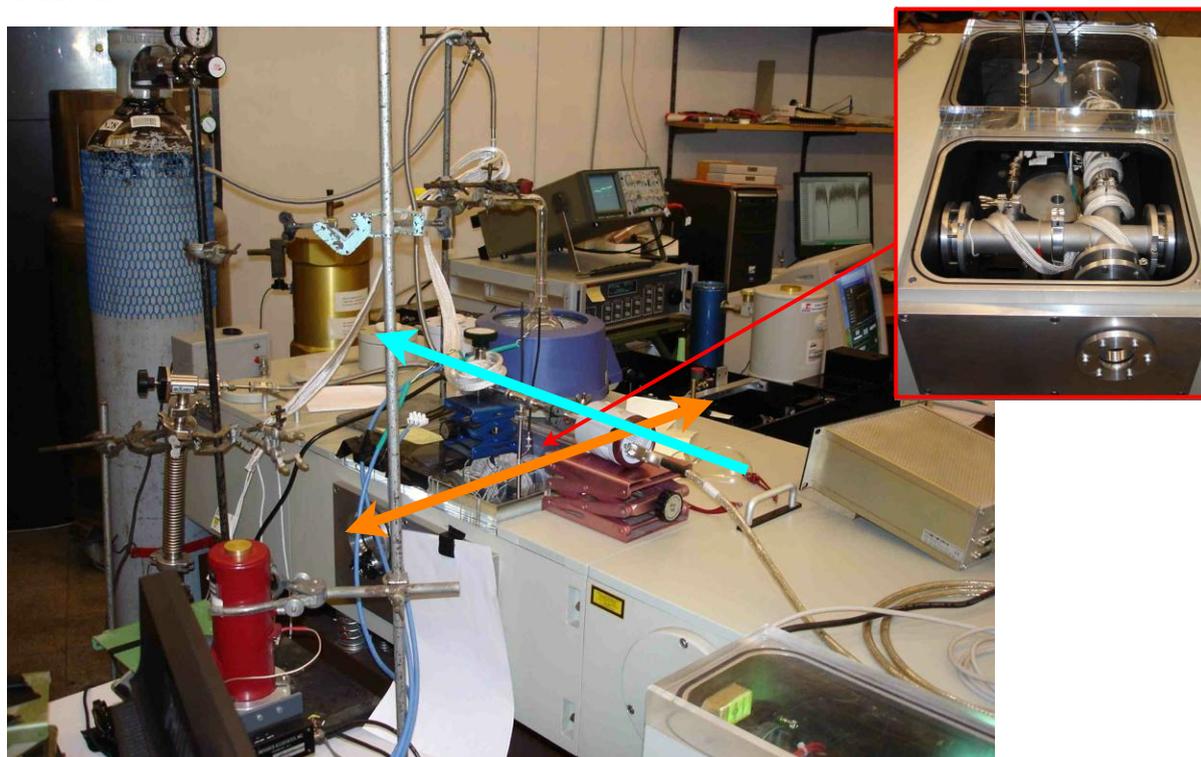


Figure 2. View of the laboratory at ULB during the simultaneous recording of FIR and IR spectra of the same sample of formaldehyde using a Bruker IFS125HR FTS and a TDL spectrometer, respectively. The cross-shaped stainless steel cell containing the sample and located inside the evacuated FTS is shown in the inset. The FIR and IR beams are represented by cyan and orange arrows (double passing in the cell), respectively.

Task 2.2.2 Re-analyse the data for C₂H₄, C₂H₆ and H₂CO with new spectroscopic data at Jungfraujoch and La Réunion.

See Task 2.1

Task 2.2.3 Intercomparison of H₂CO results from FTIR and MAXDOAS. Correlations with other biomass-burning gases

H₂CO FTIR data from Jungfraujoch have been sent to BIRA-IASB. Very preliminary comparisons with the MAXDOAS data at Jungfraujoch have been made, but they need to be refined.

Task 2.4 Production of the CCl_y and CF_y budgets for Jungfraujoch.

Although this task is supposed to start on year 3 of AGACC-II, we have already performed first investigations for the organic chlorine budget. Mahieu et al. (2011a) have combined total columns of chlorinated source gases currently available from the Jungfraujoch database, i.e. including the new CCl₄ product. At present, the CCl_y includes the following contributions:

$$[\text{CCl}_y]^* = 3 \times [\text{CCl}_3\text{F}] + 2 \times [\text{CCl}_2\text{F}_2] + [\text{CHClF}_2] + 4 \times [\text{CCl}_4]$$

Two significant contributors –and AGACC-II targets– are currently missing: CH₃Cl and CFC-113. Altogether, the four species retrieved from Jungfraujoch observations correspond to ~72% of the total CCl_y budget for the year 2004.

The CCl_y* trend over the last decade shows a constant decrease at rates close to $-(0.2 \pm 0.05)\%/yr$. It is important however to realize that the largest negative contribution of CFC-11 is currently balanced by the steady accumulation of HCFC-22.

WP 3 - Aerosol properties and radiative forcing at Ukkel

Task 3.1 Improve algorithms for aerosol characterization from ground-based spectral measurements

Task 3.1.1 Develop a better cloud screening method for deriving the AOD at 340 nm from the Brewer measurements.

This task is completed. During the previous AGACC project (SD/AT/01B), a method was developed that allows the retrieval of Aerosol Optical Depth (AOD) at 340nm from sun scan measurements of Brewer#178 at Ukkel. At that time, an initial cloud screening algorithm was developed to remove cloud perturbed AOD measurements from the results. This algorithm consisted out of 3 steps.

First, all AOD values larger than 2 were removed. Then, it was verified whether there was a direct sun observation within minutes of each individual AOD measurement. Finally, the measured irradiances (photon counts) were plotted for days with AOD measurement(s) larger than 1.5. If the graph showed clear signs of cloud perturbation, the measurement was removed. The first two steps of this cloud screening were done automatically, whereas the last step had to be done manually.

Analysis of the cloud screened data indicated that the performance of this screening technique was not optimal. Therefore it was decided to develop an improved cloud screening method. This new cloud screening method makes use of sunshine duration data (from 4 pyrheliometers at Ukkel) and is also based on the assumption that the variability of the AOD in the course of one day is either lower than 10% or lower than 0.08 AOD units. **Figure 3** gives a schematic overview of the improved cloud screening technique. The advantages of this new method are (1) the removal of the arbitrary maximum level of AOD values, and (2), it runs completely automatic (whereas the old one needed manual verification afterwards).

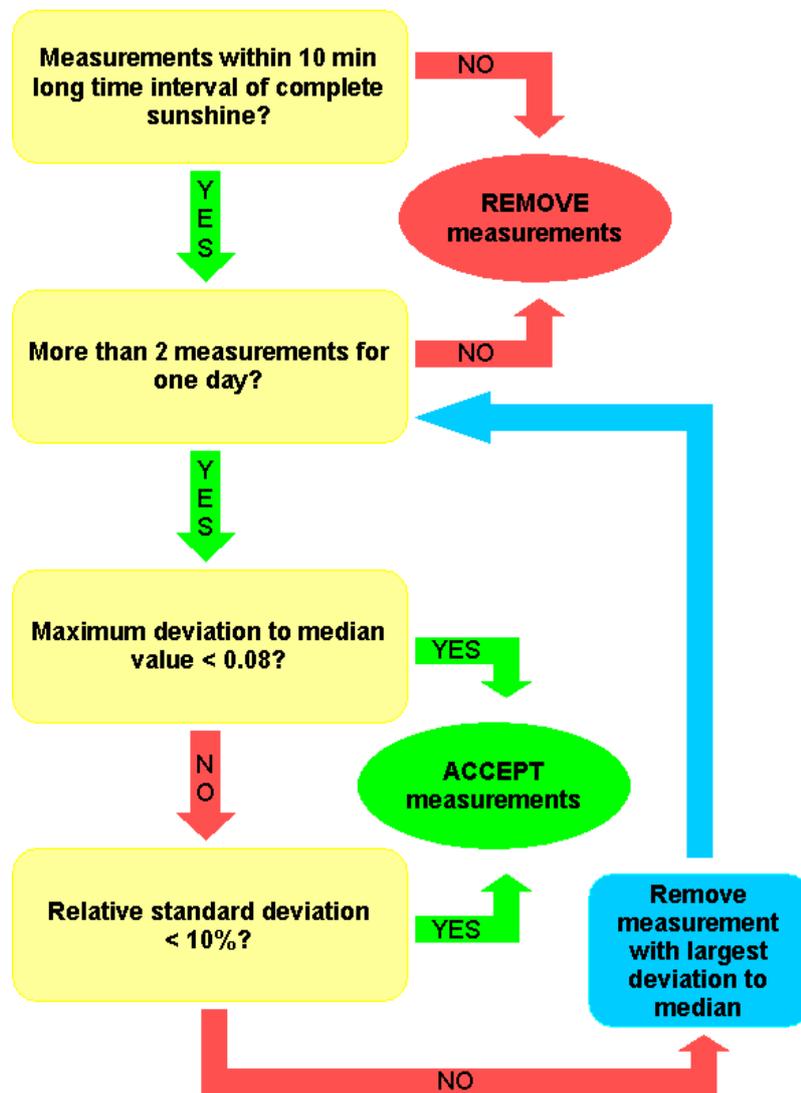


Figure 3. Overview of the improved cloud screening method.

Task 3.1.2 Develop an inverse modeling system to extract aerosol single scattering albedo information from Brewer data.

Due to the delay in the development of the new cloud screening algorithm, little progress has been made so far in the development of a method to derive the Single Scattering Albedo (SSA). There was little sense in starting the work on this task, as long as the cloud screening method was not optimal, since we need reliable AOD values to be able to derive SSA values.

Task 3.2 Operate ground-based remote sensing instruments for aerosol characterization at Ukkel

Task 3.2.1 Continue the routine operation of the Brewer spectrometer and the CIMEL sunphotometer at Ukkel, the latter in compliance with the AERONET requirements.

This (continuous) task is done according to schedule.

The data of both Brewer instruments are continuously stored and regularly checked. The Brewer data are used to build an extensive time series of AOD measurements (for Brewer016 from 1984 up to now and for Brewer178 from 2001 up to now).

In November 2011, a campaign was organised to measure the cosine response error of Brewer178 (in collaboration with Mr. David Bolsée of BIRA-IASB). The campaign proved to be a great success since the newly obtained cosine response function is much better than the one that was provided by the manufacturers of the instrument (**Figure 4**).

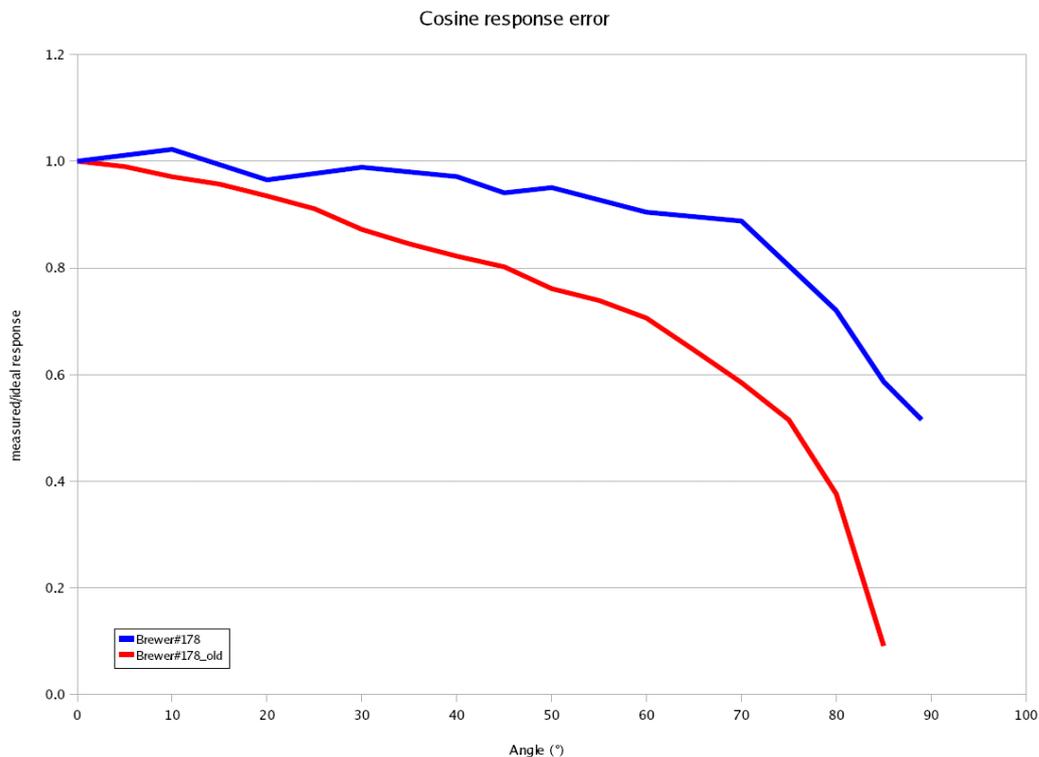


Figure 4 Results of the cosine response measurement campaign of Brewer#178. The red line is the cosine response function, provided by the manufacturers of the instrument. The blue line is the cosine response function measured during the campaign.

The CIMEL sunphotometer has been operational the whole year round, except for the period between Sept. 15, 2011 and December 9, 2011: because of a technical problem that occurred on Sept. 15, 2011, the measurements had to be interrupted, and we decided to do the calibration at the same time as the repair (in collaboration with LOA, Lille).

During the whole measurement period, the so-called cloud mode was added to the measurements on demand of B. Holben (PI of AERONET).

Task 3.2.2 Operate an aerosol lidar (ceilometer) at Ukkel and derive the aerosol backscatter ratio profiles and additional parameters (PBL, cloud information)

For this task, we are on schedule. The lidar instrument was installed in the beginning of May 2011. Every 6 seconds, information on the height of the cloud layers is provided. It also generates the backscatter profile (**Figure 5**). In the frame of the STCE project an algorithm will be implemented to derive the height of the mixing layer from these data.

Vaisala CL51 ceilometer backscatter signal 2011 09 17 (Ukkel)

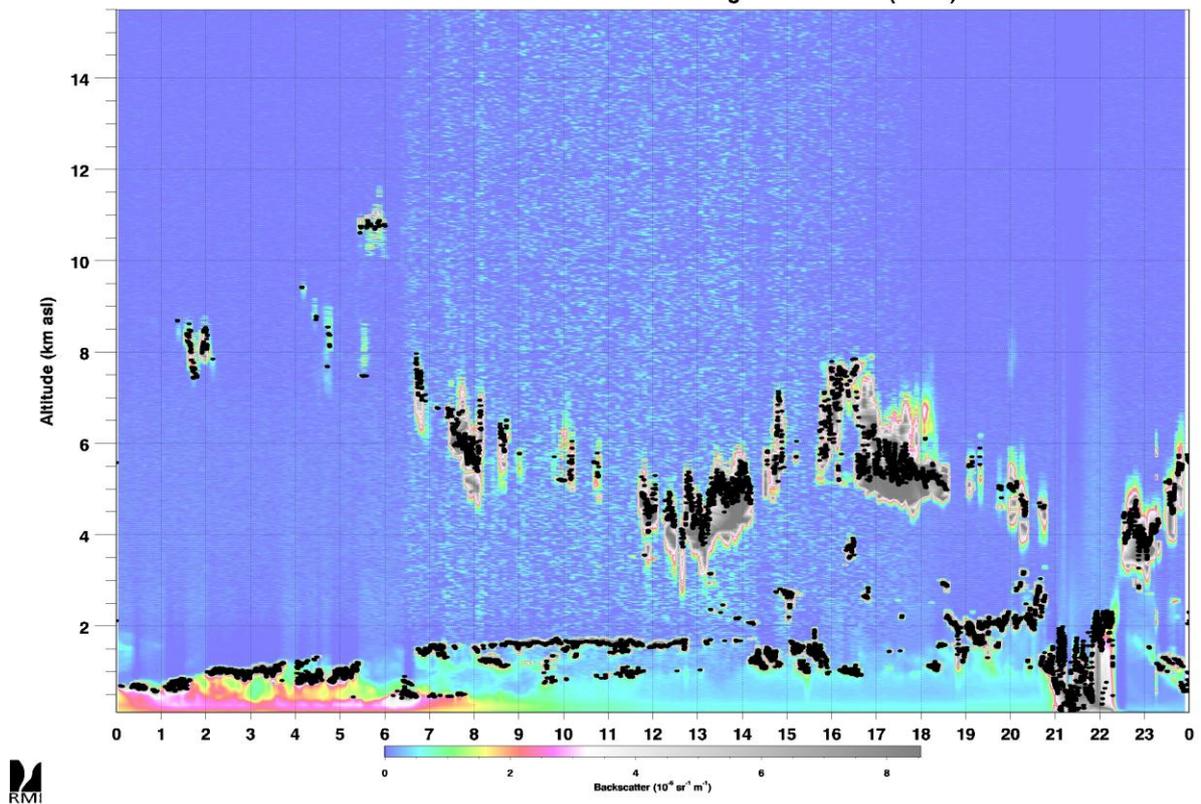


Figure 5. Example of one day of backscatter profiles measured by the Vaisala CL51 ceilometer (colour scale) and cloud base detection (black dots).

4. INTERMEDIARY RESULTS

WP1 - exploitation of FTIR spectra to derive useful geophysical information for a suite of GHGs (CO₂, CH₄, N₂O, CF₄, CCl₄)

Task 1.1. Produce time series for the major GHGs CO₂, CH₄ and N₂O

Task 1.1.1 Analyse historical grating spectra at the Jungfraujoch between 1976 and 1989, merge them with more recent data and derive long-term trends of the major greenhouse gases CO₂, CH₄ and N₂O above the Jungfraujoch.

Preliminary results for long-term time series for CH₄ and N₂O above Jungfraujoch, combining in a consistent way the data from three successive instruments, have been obtained.

Figure 6 shows the present long-term time series of nitrous oxide (N₂O) combining all total columns derived from the grating, homemade FTS or Bruker spectra. The following trends are observed: N₂O increased at a rate of (0.39 ± 0.02) %/yr from 1983 to 1995, and (0.24 ± 0.01) %/yr from 1996 to 2011 (2-sigma) indicating that the rise in N₂O is slowing down in the last years.

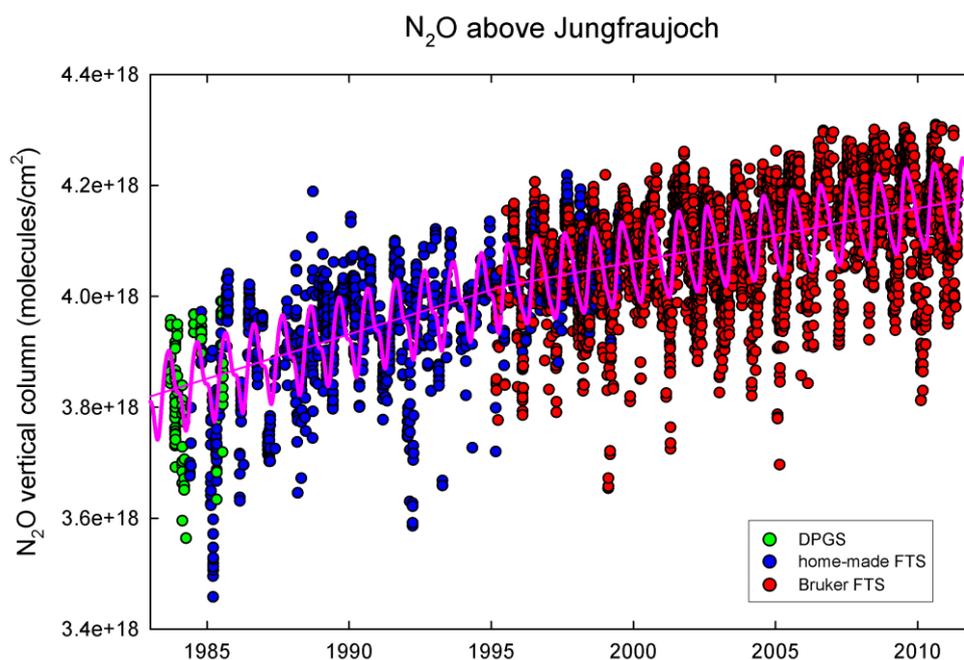


Figure 6. Total column of nitrous oxide N₂O above the Jungfraujoch, derived from spectra acquired by 3 different instruments: the 7.2-m grating spectrometer from 1983 to 1989 (green), the home-made FTS from 1985 to 2006 (blue), and the Bruker FTS from 1990 to 2011 (red). The magenta lines and curves reproduce the long-term trends (linear function with a 3rd order Fourier series) derived from the combined data set: N₂O increased at a rate of 0.39 ± 0.02 %/yr from 1983 to 1995, and 0.24 ± 0.01 %/yr from 1996 to 2011 (2-sigma).

Task 1.1.2 Implement TCCON-compliant measurements at Ile de La Reunion for the measurement of CO₂, CH₄ and N₂O.

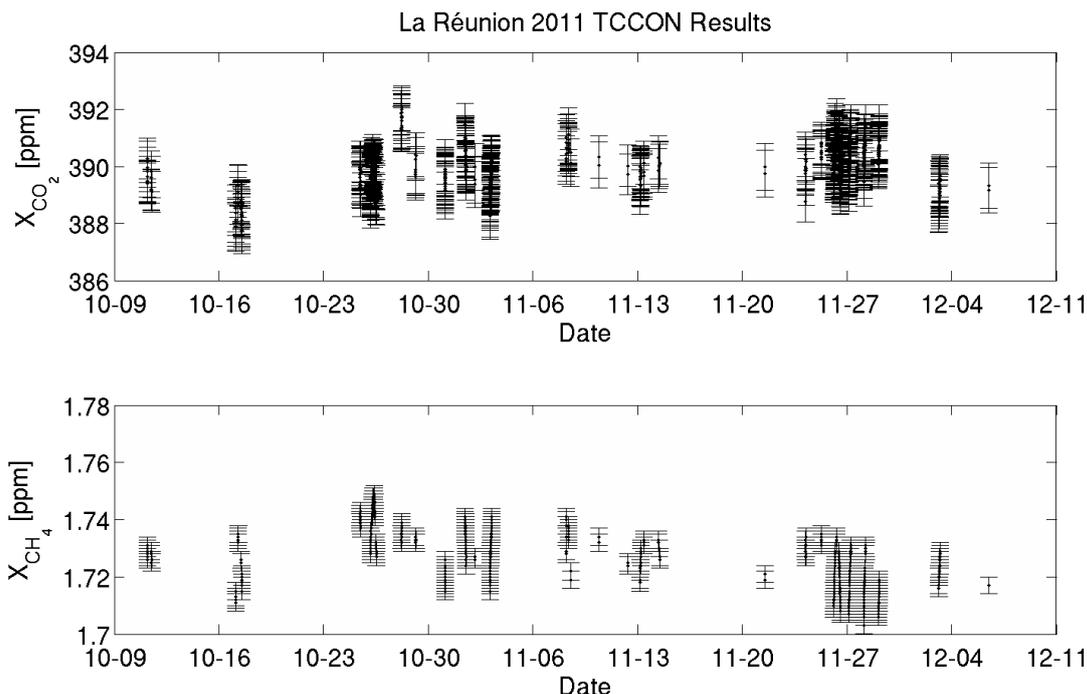


Figure 7. Time series of XCO₂ (upper plot) and XCH₄ (lower plot) observed at Ile de La Réunion since September 2011. The error bars indicate the total random error on the retrieved data.

Figure 7 shows the timeseries of column averaged volume mixing ratios of CO₂ and CH₄ (annotated XCO₂ and XCH₄, respectively) observed in 2011 at St Denis, Ile de La Réunion, since the start of the TCCON compliant measurements in September 2011.

At present, we estimate the precision of the XCO₂ measurements at 0.77 ppm or 0.2%, i.e., we achieve the precision required in TCCON. For CH₄ we achieve approximately the same precision of $0.003/1.7 = 0.2\%$.

However our dependence on the solar zenith angle is still too high, and we are investigating how to improve this.

Task 1.2 Investigate additional GHGs: CCl₄ and CF₄

Carbon tetrachloride (CCl₄)

The carbon tetrachloride (CCl₄) molecule, emitted at the ground, has been and remains a key component of the stratospheric chlorine budget, still contributing over 10 % to the total Cl loading, thus to the stratospheric ozone depletion by a similar percentage. It is also a potent greenhouse gas with a global warming potential relative to CO₂ of 1400 on a 100-year horizon. Monitoring its atmospheric budget evolution remains, therefore, of relevance to both the Kyoto and the Montreal Protocols.

All relevant Jungfraujoch spectra recorded between January 1999 and June 2011 have been systematically analyzed to produce a consistent CCl₄ time series. A fit to the CCl₄ daily mean total column data set shows a statistically-significant long-term trend of $(-1.49 \pm 0.08 \times 10^{13} \text{ molec./cm}^2)/\text{yr}$ at the 95 % confidence level. This corresponds to an annual decrease of (-1.31 ± 0.07) pptv for the mean free tropospheric volume mixing ratio, still at the 95 % statistical confidence level. These values are in excellent agreement with trends derived from *in situ* samplings in the Northern hemisphere (WMO2011, chapter 1). The negative trend of the CCl₄ loading reflects the continued impact of the regulations implemented by the Montreal Protocol and its strengthening amendments and adjustments.

A manuscript reporting about these findings has been submitted to JQSRT. It is currently under review.

Carbon tetrafluoride (CF₄)

Carbon tetrafluoride (CF₄, also known as tetrafluoromethane or PFC-14) is among the halocarbon compounds targeted by the Kyoto Protocol. It is indeed a strong greenhouse gas (GWP of 7390) further characterized by a very large atmospheric lifetime of more than 50000 yrs. CF₄ emissions have significantly increased during the last decades of the previous century. The main anthropogenic source is the primary production of aluminum while the only identified natural sources are lithospheric emissions.

All relevant Jungfraujoch spectra available from 1990 onwards have been analyzed for CF₄. After careful selection, we retained 2027 spectra recorded over 686 days.

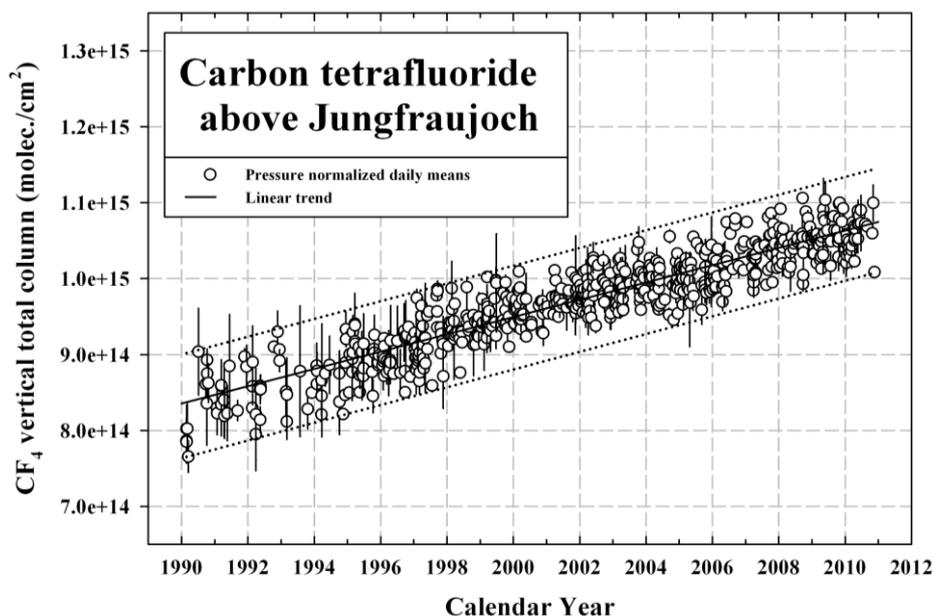


Figure 8. Long-term evolution of carbon tetrafluoride above the Jungfraujoch station (Duchatelet et al., 2011).

The resulting daily mean time series is reproduced in **Figure 8**. We determined a mean linear increase of $(1.14 \pm 0.04) \times 10^{13}$ molec./cm²/yr (2-sigma). This corresponds to a global accumulation rate of (13.2 ± 0.4) Gg/yr between 1990 and 2010. A more detailed analysis even allows distinguishing between two successive regimes: (15.4 ± 1.3) Gg/yr and (11.6 ± 0.8) Gg/yr, for the 1990-2000 and 2001-2010 time periods, respectively. It is interesting to note that these emission rates are in good agreement with those derived from *in situ* AGAGE measurements. Both techniques therefore confirm the slowing down in the accumulation of CF₄ in the atmosphere, attributed to efforts undertaken by the aluminum industry to limit its emissions despite increasing Al production. Nevertheless, CF₄ is still on the rise and considering its large warming potential and very long lifetime, this remains an important issue in the context of the Earth's global warming.

A manuscript summarizing these findings is in preparation for submission before summer.

WP 2 - Volatile Organic Compounds (VOC) and CFC-substitutes in the troposphere

Task 2.1 Develop retrieval strategies for methanol (CH₃OH) and methylchloride (CH₃Cl) at Jungfraujoch and La Réunion, and analyse the available time series. Model and satellite comparisons will be performed if possible.

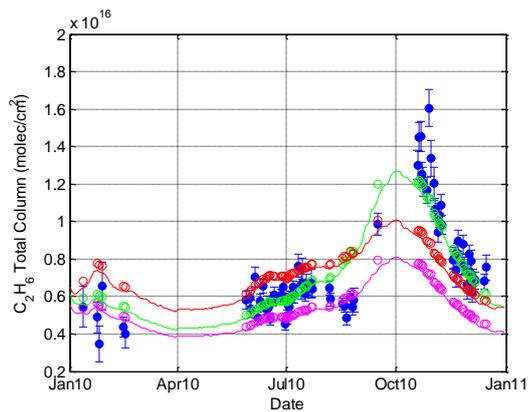
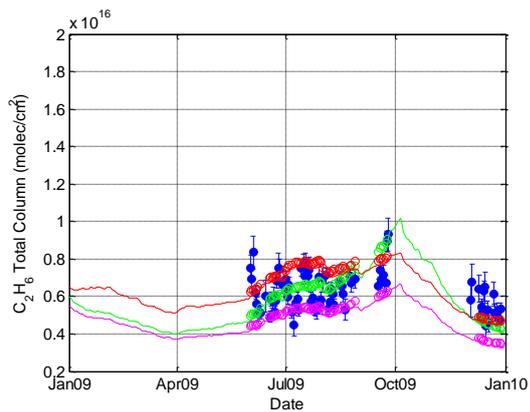
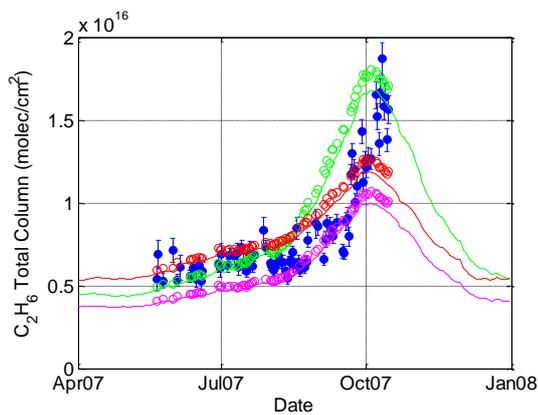
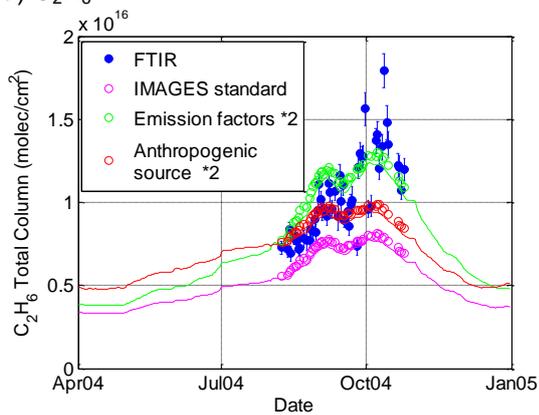
and

Task 2.2 Re-analysis of C₂H₄, C₂H₆ and H₂CO data at Jungfraujoch and La Réunion with improved spectroscopic data.

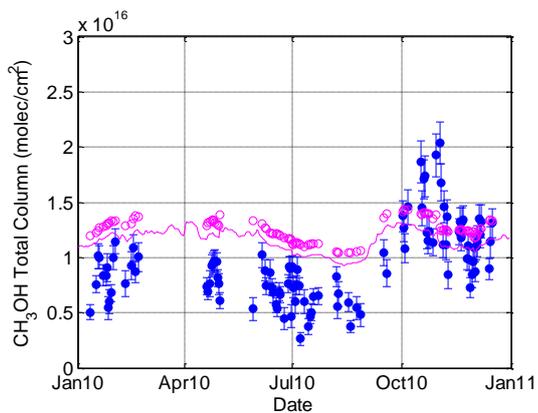
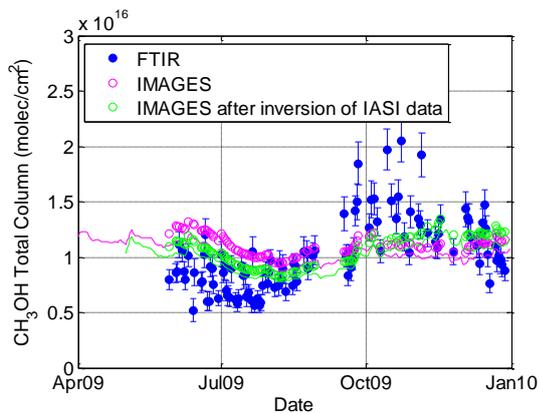
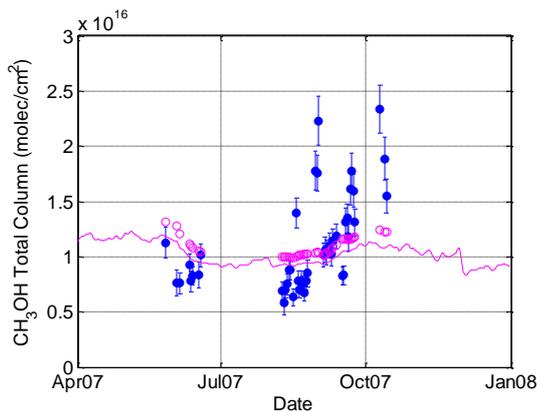
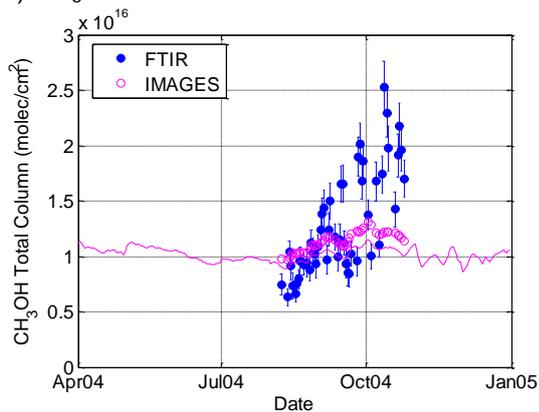
Methanol (CH₃OH) and other VOC at Ile de La Réunion

The time-series of methanol and ethane, together with HCHO, have been obtained for three FTIR measurement periods (the 2004 and 2007 campaigns and the quasi-continuous measurements in the period May 2009-2010), and compared to the CTM IMAGESv5 (**Figure 9**). One more year of measurements is available for 2011, but the model data are not yet available for this year. The seasonal cycles of the target species are nicely observed and are in agreement with the model, except for the amplitude which is in general smaller for the model.

a) C₂H₆



b) CH₃OH



c) HCHO

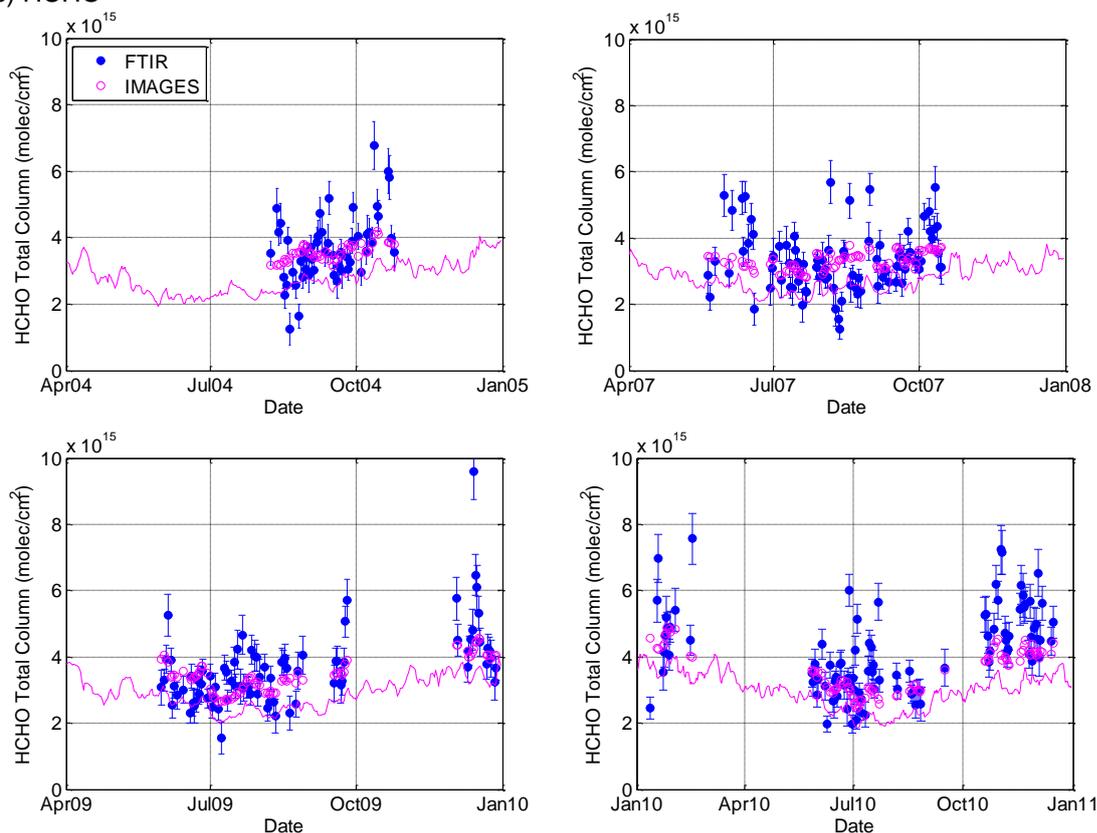


Figure 9. Daily mean total columns of C_2H_6 (a), CH_3OH (b), and HCHO (c) at Reunion Island.

The methanol FTIR total columns have already been used for comparisons with IMAGESv5 in Stavrakou et al. (2011), where the authors optimized the methanol emissions by inversion of satellite (IASI) data.

A paper about VOCs, including methanol and C_2H_6 , measured by FTIR at Reunion Island and compared to CTMs, is almost ready for submission to a peer-reviewed journal in March 2012.

Task 2.2.1 Provide improved laboratory spectroscopic data for $^{12}C_2H_4$, $^{12}C_2H_6$ and $H_2^{12}C^{16}O$

Task 2.2.1a Provide improved laboratory spectroscopic data for $^{12}C_2H_4$

We measured the intensities of about 1000 lines of the ν_7 band in two of the spectra of the first measurement set (pressure = 1.6 and 2.5 hPa). These measurements were carried out using standard techniques, i.e. by adjustment of a synthetic spectrum to the observed spectrum of a limited number of lines using a Levenberg-Marquardt least-squares fitting algorithm, assuming a Voigt profile for the lines and including instrumental contributions (Vander Auwera, 2000). Line intensities were also retrieved from four of these spectra using a multi-spectrum fitting algorithm available in Reims (Plateaux et al, 2001). Line intensities retrieved using both algorithms agree very well. However, discrepancies with the line intensities available in HITRAN (Rothman et al, 2009), varying linearly with wavenumber, are observed : line intensities measured in this work are larger than HITRAN values by about 8 % at 860 cm^{-1} and lower by about 2 % at 1080 cm^{-1} . We are investigating the origin of these discrepancies.

The second set of spectra was recorded to improve the modeling of the energy levels structure, of utmost importance to properly predict line intensities. We are presently measuring the positions of weak to very weak lines in the 3 spectra recorded.

Task 2.2.1b Provide improved laboratory spectroscopic data for $^{12}C_2H_6$

The positions of 572 lines belonging to the ν_7 band and to two other, weaker bands could be fitted only with a quite large standard deviation of 0.018 cm^{-1} (Lattanzi et al, 2011). This unsatisfactory result highlights the still too approximate character of the model. A list of parameters was nevertheless generated for 4969 lines belonging to the 5 bands included in the analysis, in the range 2900–3071

cm⁻¹. As could be expected from the rather poor fit, spectra predicted using this linelist exhibit large discrepancies with observed spectra.

Task 2.2.1c Provide improved laboratory spectroscopic data for H₂¹²C¹⁶O

About half of the IR spectra have been analyzed, relying on the line intensities reported by Perrin et al (2009) to determine the partial pressure of H₂CO. These measurements showed that the amount of formaldehyde in the cell was decreasing with time: a reduction of about 10–15 % was measured over the time required to record the FIR spectra. This relative time evolution of the H₂CO amount is needed for the analysis of the FIR spectra. A preliminary analysis of one FIR spectrum led to the determination of the partial pressure of formaldehyde in one of the experiments conducted. These preliminary results show a general good agreement between the partial pressures of formaldehyde determined (*i*) from infrared line intensities reported by Perrin et al (2009) and (*ii*) from line intensities measured in the pure rotation spectrum. However, some inconsistencies were observed in the line intensities reported by Perrin et al (2009).

WP 3 - Aerosol properties and radiative forcing at Ukkel

Task 3.1 Improve algorithms for aerosol characterisation from ground-based spectral measurements

The AODs obtained with the improved cloud screening method are compared to quasi-simultaneous CIMEL sunphotometer level 2.0 values (with a maximum time difference of 3 minutes). Due to the delays in the availability of the CIMEL data, the period of comparison is limited to a period from 1 September 2006 until 1 September 2010. For this period, 665 individual AOD values were compared and the correlation coefficient, slope and intercept of the regression line are 0.9760, 0.9816 +/- 0.0085 and 0.0776 +/- 0.0030 respectively (see also **Figure 10**). This shows that the cloud screened Brewer AOD agree very well with the CIMEL data.

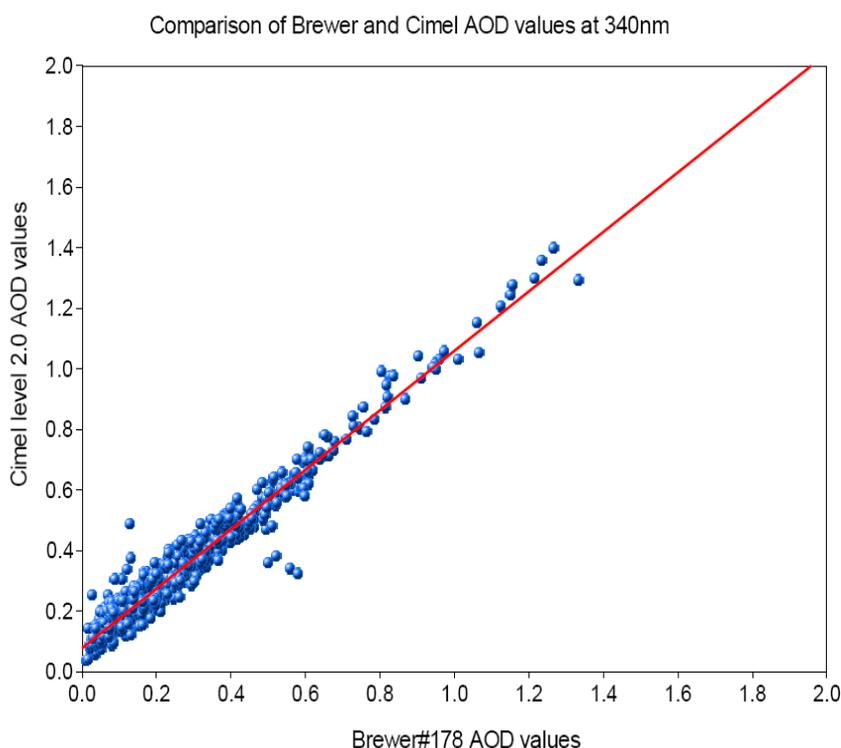


Figure 10. Comparison of the cloud screened (improved method) Brewer and CIMEL AOD values at Ukkel at 340nm.

We also investigated the temporal variability of the cloud screened AOD. The highest AOD values occur in spring, whereas in autumn and winter, the AOD are clearly lower (**Table 1**).

	2006	2007	2008	2009	2010	2011
January	-	-	-	0.23	-	0.26
February	-	0.34	0.13	0.31	0.09	0.14
March	-	0.39	0.21	0.27	0.21	0.3
April	-	0.43	0.6	0.51	0.35	0.46
May	-	0.3	0.23	0.36	0.2	0.25
June	-	0.25	0.33	0.35	0.34	0.24
July	-	0.25	0.25	0.42	0.23	0.26
August	0.13	0.17	0.24	0.22	0.15	0.22
September	0.27	0.32	0.28	0.32	0.28	0.25
October	0.18	0.16	0.2	0.22	0.12	0.14
November	0.2	0.13	0.17	0.06	-	0.14
December	-	-	-	-	-	-

Table 1. Monthly mean AOD at 340nm at Ukkel (the highest values are shown in red, whereas the lowest values are shown in blue).

Task 3.3.4 Improve UV index forecast (extra)

We spent 0.25MM on making some adjustments and improvements to the UV index forecast. The information on the website (<http://www.meteo.be/meteo/view/nl/522044-Uv.html>) was adjusted so that the maximum predicted UV index value agrees with the maximum value in the graph. (This was not the case before the adjustments.) Also, the climatological AOD, which are currently used for the UV index prediction are calculated for an extended period, from 1984 to 2010.

WP5 - Outreach

Task 5.1 To assess the results with the Follow-Up Committee. To disseminate the results in the scientific community.

A Webpage has been set up: <http://agacc.aeronomie.be>. It will be maintained in the course of the project.

Task 5.1.1 Dissemination of the results via publications, workshops, international symposia

All partners have participated to several conferences/workshops/symposia in 2011 to present AGACC-II results, e.g., the EGU-2011 General Assembly in Vienna, and the NDACC symposium in St-Denis (Ile de la Réunion).

A complete list of relevant publications and communications is given in Section 9.

Task 5.1.2 Database submissions

The Brewer ozone and UV data are deposited in the WOUDC database (woudc.org).

Task 5.2 To communicate the results beyond the scientific community: belspo, the public and policy makers

Vigouroux, C.: co-author of chapter 2 (Stratospheric Ozone and Surface Ultraviolet Radiation) of the WMO WMO/UNEP "Scientific Assessment of Ozone Depletion: 2010", published in 2011.

5. PRELIMINARY CONCLUSIONS AND RECOMMENDATIONS

The project has progressed well, and most activities seem to be on time, except for the MAXDOAS – related activities – because of problems in hiring personnel. We hope to catch up with the latter delays.

It is also worth mentioning that the CIMEL sunphotometer data at Ukkel have been included in two papers looking at aircraft observations of volcanic ash during April-May 2010 around northern Europe. In these papers, the authors have compared aircraft derived aerosol size distributions against AERONET retrievals from 3 sites (Cabauw, Brussels and Hegoland) on 17 - 18th May when a large plume of ash descended through the north sea with clear skies (**Figure 11**).

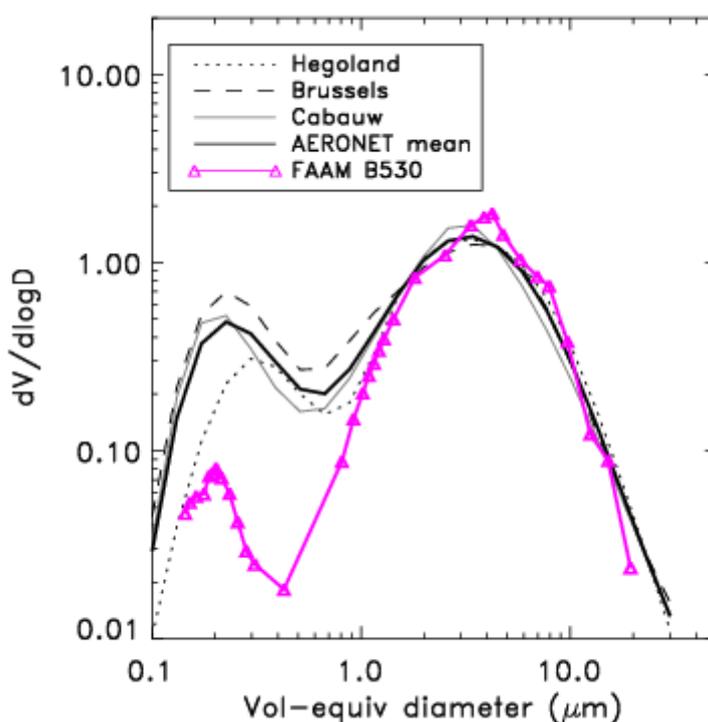


Figure 11. Normalized aerosol volume size distributions ($dV/d\log D$) from mean PCASP & CAS on flight B530 (17 May) and mean AERONET retrievals for 17 – 18 May from selected sites (see section 6.3), plus the mean from all three sites.

Status of AGACC-II deliverables expected in year 1 of the project:

- **D1.4** [M6] Technical report about retrieval strategies for CF_4 and CCl_4 at Jungfraujoch and
- **D1.5** [M12] Timeseries and trends for CF_4 and CCl_4 at Jungfraujoch
 - o A manuscript reporting about the retrievals and findings for CCl_4 has been submitted to JQSRT. It is currently under review.
 - o A manuscript reporting about the retrievals and findings for CF_4 is in preparation for submission in summer.
- **D5.1** [M6-48] Scientific publications in peer-reviewed journals, in conference proceedings; posters and oral communications at symposia
See section 9
- **D5.4** [M6 - 48] AGACC-II Webpage
See <http://agacc.aeronomie.be>

6. FUTURE PROSPECTS AND PLANNING

Due to some problems (e.g. in acquiring the necessary sunshine duration data), we spent more time (+2.5 MM) developing the new cloud screening than we originally intended to do. This time was taken from task 3.1.2. But since we were able to finish this task in 2011, the foreseen 2.5MM (between Jan and Jul 2012) can be spent on task 3.1.2 (develop an inverse modeling system to extract aerosol single scattering albedo information from Brewer data)

We hope to catch up with the delays in the MAXDOAS-related activities in 2012.

The planning of the measurements campaign in Burundi (Bujumbura) is proceeding well: the head of the Physics Department of Bujumbura University has visited BIRA in February 2012 for discussing the logistics of the installation of a MAXDOAS instrument in Bujumbura in 2012, and the scientific collaboration between the two institutes (BIRA-IASB and the University of Bujumbura). It is planned to visit the site in April 2012 for making further agreements.

We are preparing the acquisition of an additional CIMEL sunphotometer, an aethalometer and a nephelometer for making a more complete suite of aerosol measurements in Ukkel, and possibly later in Bujumbura.

At present, it appears that all other tasks should be carried out in year 2 of the project as scheduled initially.

Upcoming deliverables in year 2 of the project

- **D1.1 [M24]** Acceptance of Ile de La Réunion as a TCCON site :

Our measurements and time series of CH₄ and CO₂ at Ile de La Réunion have been verified and approved by the PI of the TCCON network. Before formal acceptance as a TCCON site, we should demonstrate the calibration of our data. Ideally we need an aircraft flight with in-situ profile measurements above the site. We are (1) discussing with our French partners at Ile de La Réunion whether we can organize such an aircraft campaign (the big problem is funding), and (2) discussing with the TCCON PI whether there is another option to get formal acceptance as a TCCON site.

In this latter context, we have decided to implement in-situ measurements of the greenhouse gases (CH₄, CO₂, H₂O, N₂O, CO) at St Denis at Ile de La Réunion, in compliance with the ICOS requirements, something which was not really planned at the start of the project. These in-situ measurements will be implemented and verified in the first half of 2012, in collaboration with the Laboratoire de l'Atmosphère et des Cyclones (LACy) and the Laboratoire des Sciences du Climat et de l'Environnement (LSCE). The in situ measurements will support the calibration of our TCCON measurements, knowing also that the French partners will implement similar measurements at the Maito site at Ile de La Réunion, at 2 km altitude, from the 2nd half of 2012 onwards.

- **D1.3 [M24]** Technical report concerning the consistency and precision of CH₄ measurements in different spectral regions

In Task 1.1.3 we are investigating the consistency between measurements of CH₄ in the near infrared (NIR) and the mid-infrared (MIR) (2650 cm⁻¹), and we are verifying the precision obtained in the different spectral ranges.

We have already performed the campaign at Ile de La Réunion with simultaneous measurements of CH₄ in the different spectral regions. We are now analyzing the data.

This work is done in collaboration with other NDACC and TCCON PIs. A communication about the results is planned in April 2012, during the EGU conference.

At start, the investigations in the NIR were supposed to include the 6000 cm⁻¹ and the 4000-4400 cm⁻¹ regions. From discussions with NDACC and TCCON colleagues, we have concluded that it doesn't make much sense at present to include the 4000-4400 cm⁻¹ region in the study, as this region is not used commonly by any network, and therefore the result of this study will have no immediate impact on ongoing network activities and comparisons between networks.

Therefore, we propose to omit this region (4000-4400 cm⁻¹ region) from the study.

- **D2.0** [M18] Technical report about retrieval strategies for methanol and methylchloride at Jungfraujoch and Ile de La Réunion and
- **D2.1** [M24] Time series of methanol and methylchloride concentrations above Jungfraujoch and Ile de La Réunion and comparisons with satellite and model data if feasible.

This work is well underway, as reported in the previous sections

- **D2.5** [M18] Updated spectroscopic parameters for the ν_1 and ν_5 bands of $\text{H}_2^{12}\text{C}^{16}\text{OH}$ in the 3.6 μm spectral range

This work is well underway, as reported in the previous sections

- **D3.1** [M18] Report on the cloud screening method for the Brewer AOD retrieval at 340nm

The research work has been finished. The results will be reported in a future publication on SSA retrieval.

7. FOLLOW-UP COMMITTEE

Extract from the minutes of the first meeting with follow-up committee on September 29th 2011 at BIRA-IASB, Brussels.

Introduction by M. De Mazière, Coordinator

Martine De Mazière briefly explains the goal of this meeting. A half year after start of the project, the first results can be shown to and discussed with the Follow-Up Committee. Unfortunately only a few members of the Follow-Up Committee have been able to join us. She presents the members of the Follow-Up Committee.

Martine gives a short overview of the project, shows the agenda for the rest of the morning and suggests a few ideas for discussion.

Partner presentation: KMI-IRM by V. De Bock

Veerle De Bock presents the contributions of RMI.

Martine asks if the AOD measurements of the Brewer are submitted to databanks. Veerle answers that they are not yet submitted to any databanks. They will look into the possibility to submit their data. **Pierre-François Coheur** asks whether AGACC-II focuses on boundary layer aerosol and – if so – whether it is realistic to run the backtrajectories in the boundary layer. He has some doubts regarding the feasibility of determining source regions for boundary layer aerosol.

Partner presentation: ULB by J. Vander Auwera

Jean Vander Auwera presents the contributions of ULB.

Martine wonders why there is a difference between region 1 and 2-3, in figure 'Mean H₂CO amount from IR'. Jean answers that this is because there are some discrepancies in relative line intensities. Martine asks about what Corinne and her team should use for the C₂H₆ lines. Jean answers that the use of the new data set – with adapted line positions, ie observed rather than calculated – should do a better job than the pseudolines. Calculations will be improved later if the required resources are obtained.

Partner presentation: ULg by E. Mahieu

Emmanuel Mahieu presents the contributions of ULg.

Willy Maenhaut asks what is the lifetime of CF₄. Emmanuel answers that its lifetime goes up to thousands of years. Half from CF₄ is from natural origin and half from anthropogenic origin. The aluminum production industry is reducing its emissions.

Pierre-François Coheur asks if PAN is still in the list of targeted species.

Partner presentation: BIRA-IASB UVVIS team, by M. Van Roozendael

Michel Van Roozendael presents the contributions of the UVVIS team of BIRA-IASB.

Jean-François Müller asks whether the O₃ trend in the stratosphere can explain the observed NO₂ trends. Michel answers that we are now in a period of O₃ recovery. This does not seem to provide a plausible explanation.

Partner presentation: BIRA-IASB IR team by C. Vigouroux & F. Desmet

Filip Desmet presents the contributions of the IR team of BIRA-IASB to WP 1.

Corinne Vigouroux presents the contributions of the IR team of BIRA-IASB to WP 2.

Willy Maenhaut asks what the lifetime of the measured airmasses is. Corinne explains that we are measuring a mixture of airmasses of different origin, nl. From Madagascar (short-lived), Africa, S. America (long-lived), even SE Asia.

Michel Van Roozendael asks which microwindows are used for formaldehyde. The ones used at Reunion Island are not the same as the ones used at Jungfrauoch by the ULg team. Therefore the sensitivities as a function of altitude may be different.

Pierre-François Coheur wonders about the variability of emission factors of different eco-systems (savannah, forest...). Corinne answers that – as we measure a mixture of airmasses of different origin, we cannot really attribute the observed emission factors to a single type of eco-system.

Discussion with Follow-Up Committee

Willy Maenhaut thinks that derivation of aerosol properties with a MAXDOAS instrument is a quite indirect and complex method to measure aerosol properties. He asks if other groups have been using this method as well. Michel Van Roozendael admits that it hasn't been used often yet, and that is why they are very careful with the interpretation of the results. But he also reminds the advantages of using a MAXDOAS instrument.

Willy shares an anecdote about the safety of the instruments in Rwanda and fears for a lack of security in Burundi.

Martine Vanderstraeten thanks the speakers. She wonders if clear messages could be delivered to policy makers after the end of the program. She points out the great importance of the conclusions in the reports. Martine De Mazière says that lots of the work will be processed in international networks, she gives examples. Martine Vanderstraeten admits that it is difficult to have direct interaction with policy makers, but asks for clear messages in reports. Martine De Mazière asks Martine Vanderstraeten to give her feedback about her reports in order to improve them.

Martine Vanderstraeten says that GCOS draws the conclusions from the reports. There is a particular highlight on observations in developing countries. Focal Point for Belgium is Steven de Witte (KMI). Martine De Mazière suggests that Veerle De Bock and Hugo De Backer get in contact with Steven de Witte, for getting more visibility in the reports.

Martine De Mazière thanks everybody for attending and closes the meeting.

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9. PUBLICATIONS

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9.1.1 Peer review

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9.1.2 Others

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9.2 Co-publications

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9.2.2 Others

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