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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
ALARO	hybridation between ALADIN (Aire Limitée Adaptation Dynamique Développement International) and AROME (Applications of Research to Operations at MESoscale)
AOD	Aerosol Optical Depth
BER	Backscatter to Extinction Ratio
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
EEA	European Environmental Agency
ECMWF	European Centre for Medium-range Weather Forecasts
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 TRANsmission database
IASI	Infrared Atmospheric Sounding Interferometer
ICOS	Integrated Carbon Observing System
IR	InfraRed
IRWG	InfraRed Working Group
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
MLH	Mixing Layer Height
NDACC	Network for the Detection of Atmospheric Composition Change
NIR	Near-InfraRed
O3MSAF	EUMETSAT Satellite Application Facility on Ozone and atmospheric chemistry Monitoring
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
TUV	Tropospheric Ultraviolet and Visible radiation model
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

The overall objective of the project is to advance ground-based remote sensing techniques for studying atmospheric chemistry and climate. The measurements are embedded in international networks, they are complementary to satellite observations and support satellite and model validation. The associated research contributes to current international research agendas.

In the first half of the project, many tasks have already conducted to interesting results, as demonstrated by a non-negligible quantity of publications, and communications to the scientific community (Sec. 9). Also a good collaboration between the partners has taken place.

A first focus of the project is on ground-based solar absorption infrared observations of major greenhouse gases, at the Jungfraujoch (ULg) and at Ile de La Réunion (BIRA-IASB).

At the Jungfraujoch, historic data from grating spectrometers have been joined to present data from Fourier-transform spectrometers. This required thorough re-analysis of the historic data and checks on data consistency. It has resulted in consistent 3-decadal time series for CH₄ and N₂O covering the period 1983 to 2012. First trend estimated have been derived for different regimes of increase. CO₂ is the next candidate greenhouse gas for a similar analysis.

Good progress has also been made to retrieve the additional greenhouse gases CF₄ and CCl₄. But it is unfortunate that the error budgets characterizing the derived quantities are still significantly dominated by the systematic uncertainties affecting the spectroscopic data.

At Réunion Island, we have succeeded in setting up a TCCON compliant experiment. Since September 2011, high-precision data for the column-averaged dry-air mole fractions (denoted by X) of the major greenhouse gases (CO₂, CH₄, and N₂O) are collected, analysed and submitted to the TCCON data archive. The achieved precisions for XCO₂ and XCH₄ are better than 0.25% and 0.4%, respectively, under clear-sky conditions and solar zenith angles smaller than 82°. We are also working on the integration of TCCON in the ICOS infrastructure, through our participation in 2 EU FP7 projects.

A second focus of the project is the advanced detection of volatile organic compounds (VOCs) at Jungfraujoch (ULg and BIRA-IASB) and Ile de La Réunion (BIRA-IASB). Time series of many different VOC (carbon monoxide, hydrogen cyanide, acetylene, ethane, methanol, formic acid and formaldehyde) have been retrieved from the FTIR spectra, and have been compared to model simulations and satellite data. It has been found that biomass burning events in Madagascar, southern Africa and America provide a large contribution to the observed abundances of the VOCs at La Réunion, and that the fire emission databases are still indebted with large uncertainties. A particular finding is the underestimation of the budget of formic acid, indicating the need for better understanding of sources and sinks, related chemistry and more observations. Preliminary data for methylchloride above Jungfraujoch have been produced. In many cases, the spectroscopic databases for these species are still deficient. Therefore laboratory experiments have been carried out at ULB to improve the line parameters for the 3.3 µm region of ethane, the 3.4 µm region of ethylene and the 10 µm region of formaldehyde. The experimental work carried out at ULB is combined with theoretical support made available through collaborations. Despite various experimental difficulties, good progress was made and in the second half of the project, the line lists will be finalised, submitted to the appropriate databases, and used for updating the data analysis at Jungfraujoch and La Réunion.

In addition, the retrieval strategy of formaldehyde from MAXDOAS spectra was revisited, based on results from the CINDI (Cabauw Intercomparison of Nitrogen Dioxide measuring Instruments) campaign that took place in Cabauw, The Netherlands, in June-July 2009. This revised strategy will now be applied to the spectra from the MAXDOAS instrument at Jungfraujoch, and the resulting data will then be compared to the ones obtained from the FTIR observations.

The third focus of the project is a better characterization of aerosol, based on data from multiple instruments, with a demonstration at Ukkel. Several instruments were already operational (Brewer spectrometer and CIMEL sunphotometer), more could be acquired (ceilometer, nephelometer and aethalometer) and some have also been made operational, like the ceilometer and the mini MAX-DOAS at Ukkel. The ceilometer now provides backscatter ratio profiles and mixing layer height on an operational basis. Models have been implemented and prepared for supporting the interpretation of the observations, in particular CHIMERE, OPAC, and backtrajectory modeling. Also tools for evaluating the radiative forcing due to an observed aerosol load have been prepared.

KMI-IRM developed an improved cloud-screening method for the Brewer data and an initial version of an algorithm to retrieve the aerosol single scattering albedo (SSA) from the Brewer data; this latter algorithm needs some improvements. Progress was also made at BIRA-IASB with the retrieval of aerosol properties from MAXDOAS observations, in particular through the development of an appropriate empirical cloud filtering method based on the colour index. Aerosol optical depths (AODs) have been retrieved at 360 nm by applying the bePRO profiling algorithm developed at BIRA-IASB (Clémer et al., 2010) to mini-DOAS O₄ measurements at Ukkel, and compared to co-located CIMEL data, showing a reasonably good agreement.

A trend analysis has been carried out on a 21 year long time series of Brewer data of monthly anomalies of erythemal UV, total solar radiation, total ozone and AOD at 320nm; the results will be published in 2013.

We are well prepared to start a thorough interpretation of all the aerosol observations at Ukkel, using the various instruments and models, and to evaluate their radiative forcing. These aerosol investigations at Ukkel are carried out in a close collaboration between KMI-IRM and BIRA-IASB.

The fourth focus of the project is on African emissions. To enable this study, it is planned to establish an atmospheric observatory in Bujumbura, which is situated in an area under the influence of biomass burning emissions. All preparatory work has been done, including the education of a local scientist of the University of Burundi and the local construction of housing for securing the instruments. The installation and the start of the measurement programme will take place in spring 2013.

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 intends to start an atmospheric observatory in Africa, in particular in Burundi, and therefore to initiate a collaboration with the University of Bujumbura in Burundi. Africa is an undersampled region; still it is an important source of biogenic compounds and biomass burning products.

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 emission databases and 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 enables 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. IMPLEMENTATION OF THE 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. The progress is mostly situated in the development and improvement of the spectral data analysis algorithms. The targeted 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. In addition, we managed to acquire an aethalometer and nephelometer, that will be deployed in Ukkel and maybe, at some time, for a campaign in Bujumbura. 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), the Total Carbon Column Observing Network (TCCON), 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 will start atmospheric observations also in Bujumbura (3°S, 29°E) in Africa where we plan to install a MAXDOAS spectrometer and a CIMEL in early 2013.

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 greenhouse gases CH₄, N₂O and CO₂

Within the framework of *task 1.1.1*, historical observations recorded at the Jungfraujoch station with a high-resolution grating spectrometer have been re-analysed to extend total column time series of important GHGs. These observations correspond to narrow spectral intervals in the mid-infrared which were specifically selected to encompass absorption lines of gases of atmospheric interest, among which CH₄ and N₂O. More than ten thousand double-pass grating spectra were recorded from 1977 to 1989, and first important efforts consisted in their transfer from tapes to accessible modern supports, to the cautious and thorough inventory of all available observations, to the identification and solving of internal inconsistencies (e.g. mismatch between the hour and geometry of observation), to the calibration of the spectra in cm⁻¹, to the generation of a dedicated database and to the modification of our fitting chain to accept these early observations.

Strategies were then set up for the retrieval of CH₄ and N₂O, for all available observational subsets (various diffraction orders and slit widths were used, resulting in spectral resolution from about 7 to 13 mK). These subsets were then harmonized among them and with FTIR data using quasi-simultaneous total column measurements. This allowed identifying some significant bias for CH₄, probably associated with spectroscopic inconsistencies. In contrast, N₂O intercomparisons between columns derived from coincident homemade FTS and grating observations, as well as between grating spectra, showed very good agreement, with ratios of (1.00±0.01) and (1.00±0.02), respectively, allowing to directly combine all subsets.

Task 1.1.2:

The TCCON compliant measurements of CO₂, CH₄, N₂O at Reunion Island started in September 2011; the observations are made in automatic mode, with a remote control option. The TCCON measurement and data analysis strategies were implemented successfully. In 2012 we encountered a number of problems with the electronics of the Bruker FTIR spectrometer, the solar tracker and the meteorological station. As a result of these technical problems, the FTIR observatory at Reunion Island was not operational for a total of 142 days in 2012. We further encountered a number of problems with the measurements which had to be corrected during the processing or post-processing of the data. This included a problem with the clock of the computer running the measurements at Reunion Island, and problems with an aging laser tube.

Task 1.1.3:

The studies about the consistency of the measurement of CH₄ in the near- and mid-infrared are ongoing. For this task we are collaborating with the team of Ralf Sussmann, leader of the research group “Atmospheric Variability and Trends” at the Karlsruhe Institute of Technology, who is currently doing a network-wide comparison of the methane retrievals for sites running both NDACC (mid-infrared) and TCCON (near-infrared) measurements. The harmonization of the input parameters used in the NDACC retrievals, such as the choice of the vertical pressure grid, with those used by the TCCON is an important challenge in this analysis. However, we are actively collaborating with the group of Ralf Sussmann to deliver the requested inputs.

Task 1.2 Investigate additional greenhouse gases: CCl₄ and CF₄

The carbon tetrachloride (CCl₄) molecule is a key component of the stratospheric chlorine budget as well as a potent greenhouse gas, with a global warming potential relative to CO₂ of 1400 on a 100-year horizon. Its atmospheric monitoring is therefore of relevance to both the Kyoto and the Montreal Protocols.

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 at 792 cm⁻¹ affected by spectroscopic line-mixing. Neglecting this effect resulted in large systematic residuals (see frame B in Figure 3 of Rinsland et al., 2012), 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 feature. Improvement in the residuals is obvious while the retrieved columns are significantly lower by ~15%, more consistent with *in situ* measurements.

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 line parameters.

As to carbon tetrafluoride (CF₄), a very long-lived GHG, a retrieval strategy using ground-based FTIR spectra has been set up for the first time. It is based on an interval spanning 6 CF₄ features, from 1284.7 to 1285.2 cm⁻¹. During the second year of AGACC-II, this strategy has been further refined, allowing improving the random error budget. Following this approach, a two-decade time series has been produced and analyzed.

Task 1.3 Provide laboratory spectroscopic data for CO₂ and N₂O if needs emerge

ULB is in charge of responding to specific needs for reference spectroscopic information for CO₂ and N₂O put forward by the other partners. With this aim, ULB first evaluates the literature to determine whether appropriate information exists. If not and if the equipment available makes it possible, laboratory measurements are carried out to fulfill the requests.

WP2. Volatile Organic Compounds (VOC) and CFC-substitutes in the troposphere

Task 2.1 Methanol (CH₃OH) and other VOCs at Ile de La Réunion

The retrieval strategies of methanol (CH₃OH), and other VOCs (hydrogen cyanide (HCN), ethane (C₂H₆), acetylene (C₂H₂), and acid formic (HCOOH)) have been optimized at Reunion Island. The main difficulties were the weak spectral absorption signatures of the target species and the strong interferences with the water vapor signatures at this humid site. For each of the target species, preliminary retrievals of H₂O and/or isotopologues were performed. The HCN and CO FTIR total columns at Reunion Island have been compared to the chemistry-transport model (CTM) GEOS-Chem, in collaboration with D. Jones and C. Whaley from University of Toronto. The CO, C₂H₆, C₂H₂, CH₃OH and HCOOH total columns have been compared to the CTM IMAGESv2 in collaboration with J. Stavrakou and J.-F. Müller (BIRA-IASB). With both CTM models sensitivity tests have been made in order to improve our current knowledge about the target species emission budget, by comparing their outputs with FTIR measurements at Reunion Island.

Task 2.1 Retrievals of methanol (CH₃OH) and methylchloride (CH₃Cl) at Jungfraujoch and La Réunion

As to CH₃OH, the first aim was to determine which microwindow is the most suitable for the Jungfraujoch retrievals among those used thus far in the literature. Our tests showed that all lead to very consistent results and that combining them allows to significantly improving the information content. A strategy involving two large windows (992-1008.3 & 1029-1037 cm⁻¹) has been carefully set up and characterized, and systematic retrievals of methanol are ongoing. Note that at Reunion Island, only the second spectral microwindow (1029-1037 cm⁻¹) is used for CH₃OH retrievals.

There are numerous but weak features of CH₃Cl near 3 μm. Sadly, experimental spectra indicate that line parameters corresponding to many of them are missing in HITRAN, even in the most recent edition. This was also true for the ethane interferences until very recently. Fortunately, pseudoline parameters for ethane have been produced on the basis of Harrison et al. (JQSRT 2010) cross section measurements. We 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 selected windows (between about 2966 and 2968 cm⁻¹). Although the situation remains very challenging, a subset of Jungfraujoch spectra has been used to produce a preliminary CH₃Cl data set.

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 ethylene (¹²C₂H₄), ethane (¹²C₂H₆) and formaldehyde (H₂¹²C¹⁶O)

Task 2.2.1.a Provide improved laboratory spectroscopic data for H₂¹²C¹⁶O

As is usually done, line intensities reported in the literature for H₂CO rely on measurements of the total pressure to determine the particle density. Because formaldehyde polymerizes or degrades easily, the gas phase in the cell may contain various products which can bias the particle density of H₂CO thus determined. In the present contribution, we aim to derive infrared line intensities for formaldehyde using intensities of pure rotation lines, observed in the far infrared (FIR) range, and the accurately known electric dipole moment of H₂CO to determine its particle density. As a result, spectra in the infrared (IR) and FIR spectral regions were recorded simultaneously.

Task 2.2.1.b Provide improved laboratory spectroscopic data for ¹²C₂H₆

To try and improve the description of the 3.3 μm region of the ethane spectrum, we first relied on a theoretical model developed by the group of C. di Lauro (*Università di Napoli Federico II*, Naples). This model attempts to include the vibration-rotation levels predicted to be observed at 3.3 μm, taking into account the effects of the internal rotation of the CH₃ groups. However, because of the complexity of the problem, the analysis had to be limited to the ν₇ band and 5 vibrational states identified as being the strongest perturbers of the upper state of that band, some being only partially observed (Lattanzi et al, 2011). Although this work led to a better understanding of the energy levels structure at 3.3 μm, the limited information that could be extracted from the spectra resulted in an unsatisfactory description of the ethane spectrum.

This outcome and a nice recent work on the ν₇ band (Villanueva et al, 2011) initiated a second attempt. This new rotational analysis was focused on the ν₇ band, and we only relied on the observed positions of R and P branch lines (and Q branch lines when resolved) and assigned them using information from previous work (Pine & Lafferty, 1982; Pine & Stone, 1996; Lattanzi et al, 2011; Villanueva et al, 2011). These assignments were then checked using ground state combination differences.

Task 2.2.1.c Provide improved laboratory spectroscopic data for ¹²C₂H₄

To fulfill the objectives of the proposed research, the experimental work carried out at ULB is combined with theoretical support made available through collaborations with two research teams, at the *Université de Reims Champagne Ardenne* (Prof. M. Rotger, Reims) and at the *Université de Bourgogne* (Dr V. Boudon, Dijon). These groups have indeed developed a theoretical model that describes in detail the structure of the vibration-rotation energy levels of C₂H₄ (Wenger et al,

2005). Provided the parameters involved are known, such a model can predict line positions and intensities, and the temperature dependence of the latter.

The general methodology of the work follows the same line as a frequency and intensity study performed by the same groups for the ν_{12} band alone (Rotger et al, 2008). Absorption spectra of ethylene have been recorded and line positions and intensities measured at ULB. This information was then provided to the theoreticians, who determined the proper set of model parameters by fitting them to the experimental information. The best-fit set of parameters and the model can then be used to generate comprehensive lists of line positions, intensities and lower state energies, useful for the analysis of atmospheric spectra.

Task 2.2.2: C₂H₆ from Jungfraujoch

The retrieval strategy for ethane has been completely revisited, after implementation of very recent C₂H₆ line parameters (Harrison et al., JQSRT, 2010) and for CH₃Cl (Bray et al., JQSRT, 2011). Our investigations showed that combining three windows around 2976, 2983 and 2986 cm⁻¹ is now possible and that the new parameters help to significantly reduce the fitting residuals. The DOFS is also strongly improved, from 1.4 with the previous approach to 2.5 with the new one.

Task 2.2.3: Comparison between MAX-DOAS and FTIR data for H₂CO at Jungfraujoch

An intercomparison exercise of H₂CO slant columns measurements has been conducted by BIRA using MAX-DOAS data from the CINDI (Cabauw Intercomparison of Nitrogen Dioxide measuring Instruments) campaign that took place in Cabauw, The Netherlands, in June-July 2009. Nine atmospheric research groups simultaneously operating MAX-DOAS instruments of various designs were involved in this exercise. This study led to the identification of potentially important sources of errors in H₂CO DOAS analysis. As a result, a set of updated recommendations was formulated for H₂CO slant column retrieval in the 336.5–359nm wavelength range (Pinardi et al., 2012). These reference settings will be applied to the MAX-DOAS measurements at Jungfraujoch.

Task 2.3 Retrieval feasibility studies

In the framework of tasks 2.3.1 and 2.3.2, we have been seeking for signatures of PAN and two CFC-substitutes, HCFC-142b (CH₃CClF₂) and HFC-134a (CH₂FCF₃), resp. Our search for PAN in the Reunion Island spectra was not successful up to now. As to the CFC substitutes at Jungfraujoch, we were able to detect weak systematic residuals (0.5-1.0%) when assuming no HCFC-142b or HFC-134a in the atmosphere. Three windows encompassing HCFC-142b features are currently under evaluation to verify the possibility to produce a total column time series with sufficient precision.

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, by combining total columns of chlorinated source gases currently available from the Jungfraujoch database, in particular the new CCl₄ product.

WP3. 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 Aerosol Optical Depth (AOD) at 340nm from the Brewer measurements

A new cloud screening technique is developed which makes use of sunshine duration data and is 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. The advantages of the newly developed method are (1) the removal of the arbitrary maximum level of AOD values and (2) it runs completely automatic (whereas the old cloud screening procedure needed manual verification afterwards).

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

We have developed a preliminary algorithm to extract SSA values using Brewer spectrophotometer data. The algorithm is based on the comparison of measured and modelled UV irradiances. The measured values are provided by Brewer#016 (from 290 to 325nm). To model the irradiances, we use the radiative transfer model TUV version 3.0 (Madronich, 1993) and run it for cloudless days. The “SSA retrieval algorithm” consists of different steps:

- 1) Choose a UV scan measurement (the time of this scan = the time for which the TUV model is run)
- 2) Choose the wavelength at which you want to determine the SSA
- 3) Select AOD and ozone for input of TUV model (i.e. Brewer measurements of ozone and AOD (at the selected wavelength) closest to the UV scan measurement time)
- 4) Run the TUV model with an initial SSA value
- 5) Compare the modeled and measured UV irradiances at the chosen wavelength:
 - a. If the difference between the two is less than 1%: accept the SSA value + increase or decrease the SSA value + run the TUV model again + repeat step 5 until the difference becomes more than 1%
 - b. If the difference between the two is more than 1%: increase or decrease the SSA value + run the model again

Task 3.1.3: Further algorithm developments for retrieval of aerosol properties from MAX-DOAS observations

Progress has been made in the development of an empirical cloud filtering method for MAX-DOAS observations based on the colour index (CI) and O₄ absorption. It is well-known that cloudy conditions may affect the quality of the retrieval of aerosol and/or trace gas columns and profiles from MAX-DOAS observations. In this study, a cloud flagging based on the CI (defined here as the ratio of the intensity of a measured spectrum at 410 nm, relative to that at 700 nm) has been investigated. Two selection criteria ($CI_{30^\circ \text{elevation}} \geq 1.5$ and $CI_{30^\circ \text{elevation}} > CI_{15^\circ \text{elevation}} > CI_{8^\circ \text{elevation}}$, i.e. sky less blue towards the horizon) have been applied to MAX-DOAS observations made by BIRA at Beijing and Xianghe (China). They contribute to remove high AOD (>2) retrievals corresponding to cloudy conditions, resulting in a much better agreement between the frequency distribution of the AERONET and MAX-DOAS AOD retrievals at both stations. The impact of clouds on the O₄ absorption has also been investigated by comparing measured O₄ slant columns to simulated ones. This method is found to be more challenging than the one based on the CI due to the difficulty of deriving appropriate threshold values for removing cloudy scenes.

BIRA-IASB RU2 is also currently performing a comparison of two profiling methods for the retrieval of vertical distribution of trace gases and aerosols from MAX-DOAS measurements. It is an important task for the harmonization of retrieval techniques which is currently under progress within the NDACC UV-vis Working Group. The first profiling algorithm involved in this comparison is based on the well-established Optimal Estimation Method (OEM; Rodgers 2000) assuming an exponentially decreasing a priori profile. The second one uses a simple parameterization of the profile based on the boundary layer height, boundary layer partial column, a shape parameter describing the concentration variation within the boundary layer, and a free tropospheric partial column. These profiling tools have been applied to MAX-DOAS H₂CO and O₄ measurements at Xianghe. A good agreement is found between both methods in term of total column and surface concentration for both aerosols and H₂CO. The comparison of retrieved profile heights (altitude in km below which 75% of the integrated (aerosol/HCHO) profile resides) shows that the range of retrieved values is much larger for the simple parameterization approach than for the OEM-based technique which gives profile heights most of the time close to the a priori. A validation step is needed to determine which method is better.

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

The data of both Brewer instruments are continuously stored and regularly checked. The Brewer data are used to build an extensive time series of UV radiation, total ozone and AOD. We performed a trend analysis, a change point analysis and a multiple regression analysis on a 21 year long data series

of erythral UV doses, total solar radiation (from pyranometers), total ozone and AOD at 320nm (see section 4 for first results).

In November 2011, a campaign was organized to measure the cosine response error of Brewer#178 (in collaboration with Dr. David Bolsée of BISA). The campaign proved to be a great success since the newly obtained cosine response function is much better than the one provided by the manufacturers.

Both Brewer instruments were calibrated in 2012. No major problems showed up during the calibration.

The CIMEL sunphotometer has been operated continuously at Ukkel, except for the period during which it is sent to the Laboratoire d'Optique Atmosphérique (LOA) for calibration (15 September to 9 December 2011). The data are available on the AERONET site (<http://aeronet.gsfc.nasa.gov/>).

Task 3.2.2: Operate an aerosol lidar (ceilometer) at Ukkel and derive the backscatter ratio profiles and additional parameters (planetary boundary layer height, cloud information)

A Vaisala ceilometer CL 51 was installed in May 2011 at Ukkel. The instrument can be used to determine the cloud base height and the mixing layer height (MLH). Also, the Backscatter to Extinction Ratio (BER) is determined.

The cloud base height detection algorithm is based on the detection of a strong vertical gradient in the backscatter, combined with a high backscatter signal. It allows for cloud detection every 5 minutes up to 13 km. No cloud detection is possible under circumstances with precipitation or ground-based fog.

For the detection of the MLH, we use a combination of two algorithms. The first algorithm determines the height at which there is a local minimum in the (negative) derivative of the backscatter signal, which means there is a significant vertical gradient at this height. The second algorithm determines the height at which a maximum in the temporal variance of the backscatter profile (within 5 minutes) occurs. If the resulting heights of the two algorithms agree to within 165m, this value is accepted as the MLH. The detection of the MLH is not possible in circumstances with precipitation, ground-based fog, strong convective conditions or too low aerosol concentrations.

Finally, the lidar can also determine the BER by using an iterative inversion approach in which the sunphotometer measurements are used to constrain the lidar inversion. The BER is increased (decreased) if the lidar derived optical thickness is larger (smaller) than the one from the sunphotometer.

Task 3.2.3: Operate a mini MAX-DOAS instrument at Ukkel during the project

A mini-DOAS instrument has been continuously operated in Ukkel by BIRA-IASB RU2 since April 2011 (see Figure 1). The retrieval of the aerosol content based on MAX-DOAS measurement of O_4 has started and preliminary results are shown below (see Task 3.3.1). It should be noted that tropospheric NO_2 columns have been derived using the geometrical approximation, i.e. the NO_2 layer is assumed to be located below the scattering altitude at 30° elevation, so tropospheric NO_2 vertical columns can be derived by applying a geometrical AMF to measured NO_2 slant columns. This data set has been used to validate satellite observations, in particular GOME-2 data as part of the O3MSAF (EUMETSAT Satellite Application Facility on Ozone and atmospheric chemistry Monitoring).

Task 3.3 Interpretation of the aerosol data at Ukkel

Task 3.3.1: Demonstrate the strength of the aerosol remote sensing

Aerosol optical depths (AODs) have been retrieved at 360 nm by applying the bePRO profiling algorithm developed at BIRA-IASB (Clémer et al., 2010) to mini-DOAS O_4 measurements at Ukkel, and compared to co-located CIMEL data.



Figure 1: Picture of the BIRA-IASB mini-DOAS instrument installed on the roof of KMI at Ukkel and pointing towards Brussels city center.

Task 3.3.2: Study the agreement between aerosol optical properties measured with the set of instruments mentioned above and simulated with Chimere.

Chimere provides the chemical composition of aerosol particles up to a height of 5.5km. This chemical composition will be used as input for the OPAC model to get the optical properties (AOD and SSA). Before we can start with the actual modelling using OPAC, some alterations concerning the format of the output of Chimere have to be done (see section 4).

WP4. African emissions

Task 4.1 To install and operate a MAX-DOAS instrument in Bujumbura (Burundi)

A new MAXDOAS instrument has been assembled at BIRA with the aim to install it at the University of Burundi (located in Bujumbura, North of Lake Tanganika close to the border with RDC) in early 2013. This instrument is similar in concept and design to the MAX-DOAS systems already successfully operated in Xianghe (China) and at the Jungfraujoch (Switzerland) since 2010. Measurements in Central Africa will allow us to extend the ongoing monitoring of atmospheric composition at a site of high interest, due to its proximity with biomass burning and volcanic sources. Results will be used in combination with 3D-CTM and dispersion models to investigate the impact of atmospheric transport and chemistry on air quality in Bujumbura. They will also provide a very useful and unique data set for satellite validation. In the course of 2012, active contacts have been established with the University of Burundi in particular during a visit by M. Van Roozendael and C. Hermans in May 2012. This was followed in autumn by the organization of a 3-months training session at BIRA for M. Eugène Ndenzako (physicist at the University of Burundi), during which period he could contribute to the preparation of the MAX-DOAS instrument, its calibration in the lab, and initial test in the field at BIRA. At the same time he was also trained to the usage of the various control and evaluation software packages. This knowledge will enable Eugène to operate the MAX-DOAS instrument once installed at the University of Burundi in spring 2013 and also to contribute to the data exploitation in the remaining part of the AGACC-2 project (until end of 2014). Additional resources will be looked for, to possibly start a PhD.

WP5. Outreach

AGACC-II has already led to various communications and peer-reviewed publications (see Section 9). The data produced in AGACC-II have been used by other research teams, including modelling teams, teams working in the Solar-terrestrial Center of Excellence and by users of the databases to which data are submitted.

4. PROGRESS AND 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 greenhouse gases CH₄, N₂O and CO₂

First trend results have been deduced from the very long-term time series of CH₄ and N₂O above Jungfraujoch. These two data sets combine in a consistent way column data from three successive instruments. For CH₄, the trend analysis indicates four consecutive regimes, with relative yearly rates close to 0.8% over 1983-1992, 0.4% over 1993-1999, 0% over 2000-2004 and 0.4% over 2005-2010. This is in good agreement with CHASER model results, except over the last years. This points to the need for the model to include updated emissions for that period, e.g. the anomalous emission of methane from wetlands in 2007. Very good agreement is also observed when comparing with *in situ* trends from the AGAGE station of Mace Head (Ireland).

For N₂O, a more regular accumulation is revealed by our three-decadal time series. Nevertheless, we can distinguish between two regimes, with yearly rates of $(0.39 \pm 0.02)\%$ for 1983-1995 and $(0.24 \pm 0.01)\%$ for 1996-2011 ($2\text{-}\sigma$), indicating a significant slowing down of the N₂O build up over the last fifteen years. Comparison with *in situ* data is ongoing.

At Réunion Island, we are building up a timeseries of XCO₂, XCH₄ and XN₂O (XM stands for the column averaged dry-air mole fraction). The first six months of data are publicly available from the TCCON archive; newly acquired data will be made available after a delay of 3 – 6 months. For XCO₂, we achieve a precision of order 0.25% (~1 ppm).

Fig. 2 shows the actual status of the timeseries.

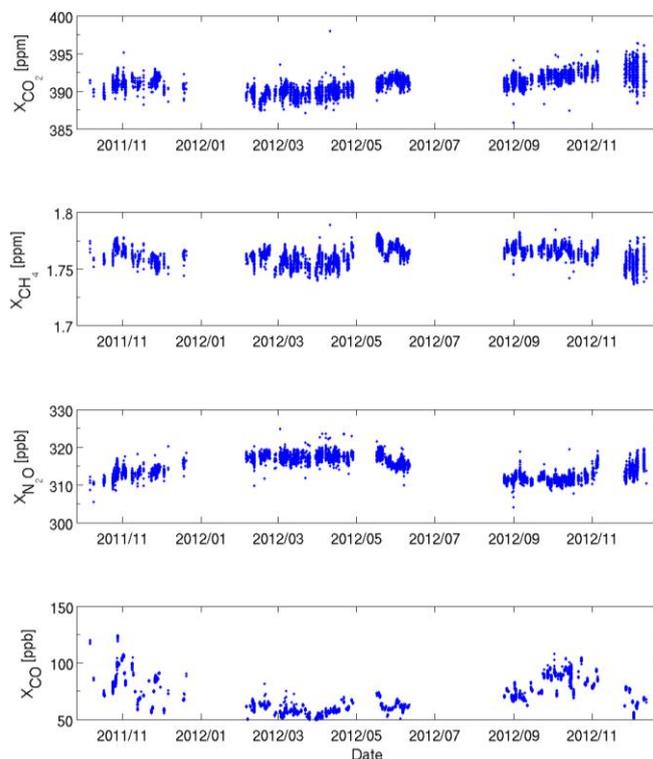


Fig. 2: Timeseries of (from top to bottom) XCO₂, XCH₄, XN₂O and XCO above Reunion Island. The increase observed in November 2011 is caused by local fires on the Island. The larger scatter in the end of the timeseries is due to instrumental problems (laser degradation).

Task 1.2 Investigate additional greenhouse gases: CCl₄ and CF₄

The study of CCl₄ has been finalized and published (Rinsland et al., 2012). The main findings are: (i) neglecting line-mixing in a strong CO₂ Q-branch interference results in CCl₄ columns biased high by $(15 \pm 2)\%$, (ii) the CCl₄ abundance decreases at $-(1.1 \pm 0.1)\%/yr$ over 1999-2012, in good agreement with

in situ observations, as a consequence of measures adopted by the Montreal Protocol, (iii) CCl₄ remains a significant contributor to the organic and inorganic chlorine budgets.

For CF₄, our most recent data set reveals a mean linear increase of $(1.14 \pm 0.04) \times 10^{13}$ molec./cm²/yr (2-sigma), or (13.2 ± 0.4) Gg/yr between 1990 and 2011. Here also, two successive regimes are identified, with emissions of (15.4 ± 1.3) Gg/yr and (11.6 ± 0.8) Gg/yr, for 1990-2000 and 2001-2011, 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.

WP2. Volatile Organic Compounds (VOC) and CFC-substitutes in the troposphere

Task 2.1 Methanol (CH₃OH) and other VOCs at Reunion Island and Jungfraujoch.

a) Reunion Island

Our retrieval strategies of HCN, C₂H₆, C₂H₂, CH₃OH, and HCOOH and the comparisons with the models GEOS-Chem and IMAGESv2 have been published in Vigouroux et al., (2012). The FTIR formic acid time-series were also used in Stavrakou et al. (2012), an emission inversion study constrained by IASI data. A similar approach was adopted in Stavrakou et al. (2011) for methanol, in which our FTIR data also contributed. The formic acid FTIR total columns were also used in Paulot et al. (2011). Furthermore, the HCN and C₂H₂ FTIR data were used for the validation of IASI products in Dufлот et al. (2012).

The main results of the comparisons with the models discussed in Vigouroux et al. (2012) are the following. First, the fire emission database used in IMAGESv2 (GFED3; van der Werf et al., 2010) seems to underestimate the pyrogenic emissions, especially in the late September-October period in south-eastern Africa - Madagascar region. Second, the excellent correlation of CH₃OH and HCOOH with CO between August and November suggests that, despite the dominance of the biogenic source of these compounds on the global scale, biomass burning is their major source at Reunion Island during this period. This was expected for other species such as HCN, C₂H₆ and C₂H₂, but less for methanol and formic acid given the low contribution of fires to their global budget (2% and 14%, respectively). We show in Fig. 3, the FTIR and IMAGESv2 simulations of methanol and acid formic. The optimized IMAGESv2 simulations, using IASI data, are shown in green in Fig. 3. We observe that the CH₃OH IASI derived emissions remain too low during the fire season, suggesting that IASI may underestimate CH₃OH in this period in the Southeastern Africa-Madagascar region.

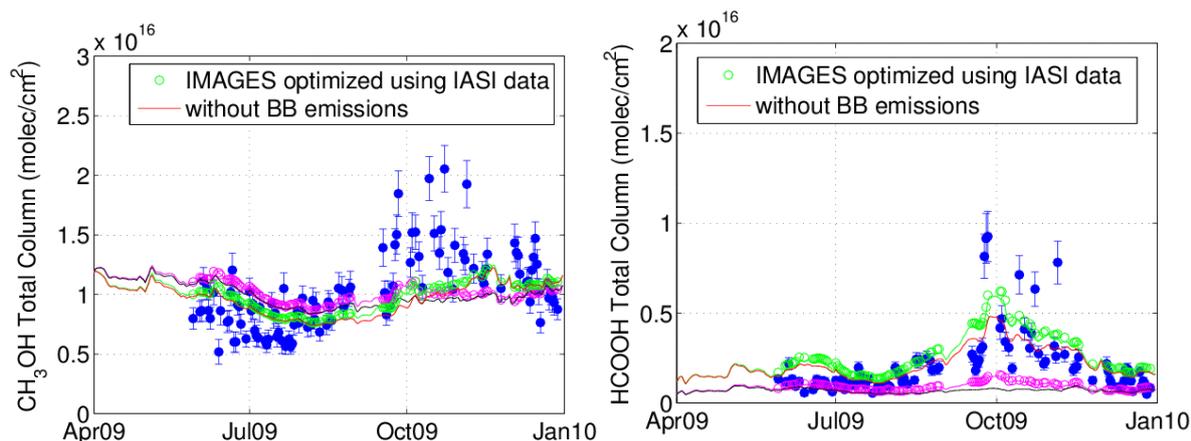


Figure 3: Time-series of daily mean total columns at Reunion Island: CH₃OH (left panel) and HCOOH (right panel). The FTIR data are represented by the blue filled circles, the IMAGESv2 model simulations by the colored lines (magenta for the standard runs; green for the optimized simulations using IASI data), and the model data smoothed with the FTIR averaging kernels with the open circles. The model simulations obtained when the biomass burning (BB) contribution is removed are shown in black for the standard run, and in red for the inversion using IASI data.

From Fig.3, we see that, although the IMAGESv2 optimization of HCOOH sources using IASI data greatly improves the agreement with FTIR data during the fire season (Fig. 3, right panel, green curve), the contribution of biomass burning given by IMAGESv2 is only minor (red curve compared to green curve). This specific result at Reunion Island seems to disagree with the conclusion based on FTIR measurements that biomass burning is a dominant source of HCOOH at Reunion Island during

the August–November period. On the other hand, the FTIR finding is consistent with the study of Paulot et al. (2011). Indeed, they have shown that a good agreement between the HCOOH columns modeled by GEOS-Chem and the FTIR measurements at Reunion Island can be achieved by assuming that organic aerosol (OA) oxidation generates a diffuse source of formic acid, knowing that the dominant source of OA in the Southern Hemisphere is biomass burning.

b) Jungfraujoch

At the moment, a methanol time series has been produced for the 1997–2011 time period. It reveals a strong seasonal modulation, with minimum values and variability in winter. In summertime, total columns are on average 4 times larger; the ratio between the lowest and largest individual columns exceeds 14. No long-term trend emerges from our analysis, in agreement with similar investigations based on Kitt Peak observations.

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

Task 2.2.1a Provide improved laboratory spectroscopic data for H₂¹²C¹⁶O

Spectra of formaldehyde were recorded at ULB simultaneously in the FIR range (0–400 cm⁻¹), using a Bruker IFS125HR FTS, and in the IR region (near 3.4 μm), using a TDL spectrometer available in the group of Prof. M. Lepère (*Facultés Universitaires Notre Dame de la Paix, FUNDP, Namur*) and installed at ULB. The formaldehyde sample (~1.5 % diluted in 10 Torr of N₂) was contained in a cross-shaped stainless-steel cell located inside the evacuated FTS. A total of six experiments were conducted at 296 K with six different formaldehyde samples. During each experiment, one high-resolution FIR spectrum and about 30 IR spectra were simultaneously recorded. The recording of the FIR spectrum was then extended overnight, to yield a high-resolution spectrum with improved signal-to-noise ratio.

The six sets of IR spectra probe five different, ~0.5 cm⁻¹ wide regions of the H₂CO spectrum. The analysis of the two sets probing the same region (2911.5–2912.2 cm⁻¹, referred to as R1 hereafter) and an additional set (2928.4–2929.2 cm⁻¹ region, R2) was carried out. Relying on the line intensities reported by Perrin et al (2009) and provided in HITRAN (Rothman et al, 2009), the analysis of these spectra showed that the amount of formaldehyde in the cell decreased by about 10–15 % over the time required to record the corresponding FIR spectra. Using this relative time evolution of the H₂CO amount, the accurately known electric dipole moment of H₂CO (Fabricant et al, 1977) and pure rotation line intensities measured in the 40–55 cm⁻¹ range, the mean partial pressure of formaldehyde was determined for these three experiments. The absolute intensity of the IR H₂CO lines probed in region R1 was then determined. These preliminary results indicate that the line intensities available in HITRAN (Rothman et al, 2009) for these lines may be too high by about 7 %.

However, we faced difficulties. If we were able to identify the first two IR regions probed by the TDL using HITRAN (Rothman et al, 2009), we could not identify the three remaining sets. Therefore, we recorded FTS spectra of the whole ν₁ and ν₅ band system (2500–3200 cm⁻¹). With their help, this problem was solved for one of these regions and we are confident that the two remaining ones will also be similarly identified. Unfortunately, these FTS spectra put forward another problem. Indeed, they showed that the relative intensities provided in HITRAN (Rothman et al, 2009) for the H₂CO lines observed in the TDL spectra agree within about 3 %. If the partial pressure determined from the analysis of the TDL spectra probing region R1 indeed yield consistent results for the two sets recorded (within 1 %), the two H₂CO lines observed in spectra of region R2 yield partial pressures differing by as much as 8 %. The origin of such a discrepancy is still unclear.

Finally, we also studied the range from 29 to 55 cm⁻¹, relying on FIR spectra recorded overnight, to investigate the possible existence of a Herman-Wallis dependence of the pure rotation line intensities. Such dependence would indeed affect the partial pressure of H₂CO inferred from the pure rotation line intensities. None could be observed.

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

We recorded four high-resolution spectra of the 3.3 μm region of ethane, using a Bruker IFS125HR FTS. In a first analysis, 572 line positions belonging to the ν_7 band and to two other, weaker bands could be fitted with a quite large standard deviation of 0.018 cm^{-1} only (Lattanzi et al, 2011). A list of parameters was nevertheless generated for 4969 lines in the range $2900\text{--}3071\text{ cm}^{-1}$. As could be expected from the rather poor fit, spectra predicted using this linelist exhibit rather large discrepancies with observed spectra.

We performed a second rotational analysis of the ν_7 band, relying on line positions observed in the recorded spectra and literature data (Pine & Lafferty, 1982; Pine & Stone, 1996; Lattanzi et al, 2011); no modelling of the vibration-rotation structure was done. We were able to extend the rotational assignments in the ν_7 band, bringing to more than 3000 the total number of assigned lines. As an example of the outcome of this second analysis, Fig. 4 compares the modelling of the $^1\text{Q}_2$ branch of the ν_7 band obtained using HITRAN (Rothman et al, 2009) with our results.

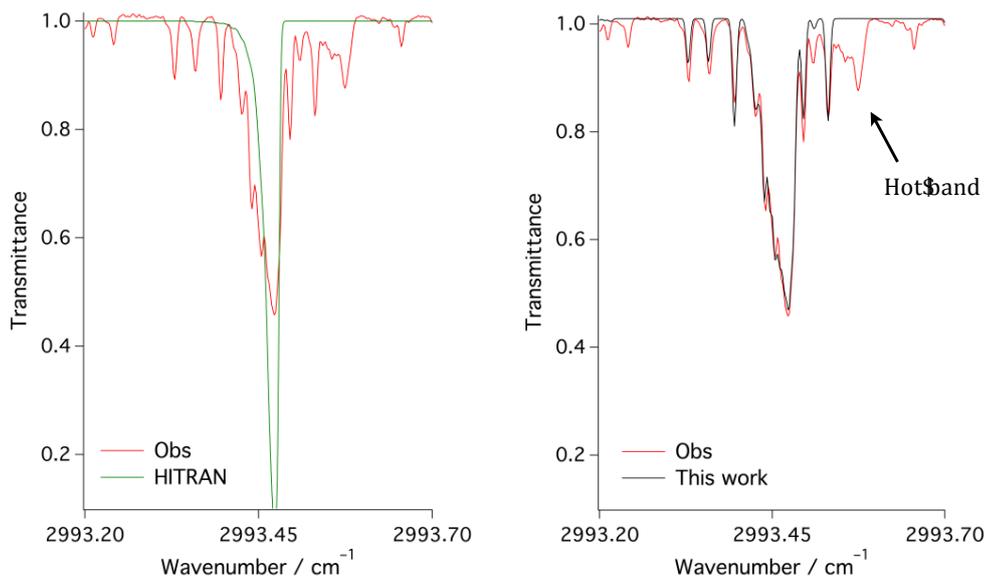


Figure 4: Observed spectrum of the $^1\text{Q}_2$ branch of the ν_7 band (280 K, 19.7 cm, 0.2 hPa) and spectra calculated using HITRAN (Rothman et al, 2009) and results of the present analysis.

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

We recorded two sets of absorption spectra of the 10 μm region using a high resolution Bruker IFS125HR FTS, one tailored to the observation of the strong ν_7 band and the other adapted to the weak ν_{10} and ν_4 bands. A combined frequency analysis of the ν_{10} , ν_7 , ν_4 and ν_{12} bands was carried out in collaboration with the French theoreticians. More than 10000 line positions, measured for the ν_{10} , ν_7 , and ν_4 bands in the spectra recorded and resulting from our previous work (Rotger et al, 2008) for the ν_{12} band, were fit together with an overall standard deviation of 0.00034 cm^{-1} . This improved frequency analysis of the 10 μm region is almost completed. Apart from the line positions and lower state energies, an important outcome of this analysis are the wavefunctions associated with the upper energy levels, essential for the modelling of the line intensities.

The intensities of about 1000 lines of the ν_7 band were measured at ULB and at the Université de Reims Champagne Ardenne, using two different computer codes. These line intensities 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 by about 8 % at 860 cm^{-1} and lower by about 2 % at 1080 cm^{-1} . We have not yet understood the origin of this discrepancy.

Task 2.2.2: Re-analyse the data for C_2H_4 , C_2H_6 and H_2CO with new spectroscopic data at Jungfraujoch and La Réunion.

The new 3-window retrieval approach has been used to produce an updated C_2H_6 data set at Jungfraujoch spanning the 1994-2011 time period. A first trend analysis indicates a slight decrease of $-(0.47 \pm 0.35)\%/yr$ for the total columns ($2\text{-}\sigma$). Model data available over the 1998-2005 time period also show a decrease: of $-(1.58 \pm 0.68)\%/yr$ for the CHASER model (Sudo et al., 2002), and of $-(0.89 \pm 0.56)\%/yr$ for the Oslo

CTM simulations (Berntsen et al., 1997). Hence, the two modelled trends agree within 2- σ and all data sets provide statistically significant negative trends for ethane. As to the seasonal modulations, the magnitude of the seasonal change is better appraised by the Oslo model while the positions of the minimum and maximum columns are better predicted by the CHASER simulation.

Task 2.4 Production of the CCl_y and CF_y budgets for Jungfraujoch

At present, the CCl_y* includes the following contributions:

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

All together, the four species currently 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 canceled by the steady accumulation of HCFC-22.

WP 3. Aerosol properties and radiative forcing at Uccle

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 340nm from the Brewer measurements

The Brewer AOD values at 340nm, obtained with the improved cloud screening method, are compared to quasi-simultaneous CIMEL sunphotometer level 2.0 data (with a max. time difference of 3 minutes). For a period from 1 September 2006 until 1 September 2010, 665 individual AOD values are 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. This shows that the cloud screened Brewer AOD agree very well with the CIMEL data. The newly developed cloud screening method has now also been applied to the Brewer AOD retrieval using direct sun measurements at 306, 310, 313, 316 and 320nm.

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

The first preliminary results of the SSA retrieval algorithm are available and have been analyzed. Currently, we have modelled SSA ranges (calculated for a time around 12UT) for 84 cloudless days between September 2006 and December 2010. From the results, it is clear that the SSA range becomes very large when the input AOD value is low as well. There is also a clear relationship between the input AOD and the mean modelled SSA value, with higher AOD values leading to higher and more reliable SSA values. SSA values modelled for AOD values below 0.5 seem to be very unrealistic.

Task 3.1.3: Further algorithm developments for the retrieval of aerosol properties from MAX-DOAS observations

This task has not progressed as expected due to the long-term absence of Dr. K. Clémer who was in charge of these aspects within BIRA-IASB RU2. A new collaborator, Dr. C. Gielen, is now working full time on MAX-DOAS aerosol activities, so we hope to catch up with the delays by the end of the project.

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

The first results of the time series analysis of monthly anomalies of erythemal UV, total solar radiation, total ozone and AOD at 320nm show that only the AOD has a negative trend over the 21 year time period ($-14.97\% \pm 4.97\%$ per decade). The trends in erythemal UV doses, total solar radiation and ozone are positive ($+7.58\% \pm 1.95\%$ per decade for UV; $+5.07\% \pm 1.52\%$ per decade for total solar radiation and $+2.88\% \pm 0.00\%$ per decade for ozone). All trends are significant at the 99% level, except for the AOD trend which is significant at 95%.

Task 3.2.2: Operate an aerosol lidar (ceilometer) at Ukkel and derive the backscatter ratio profiles and additional parameters (planetary boundary layer height, cloud information). The lidar is now fully operational and provides us with cloud base heights and mixing layer heights. For each day, figures (such as Fig. 5) are available which show both heights and which can be used, for instance, by the weather forecasters. Also, for each day, the lidar MLH is compared with the MLH from ALARO and from ECMWF.

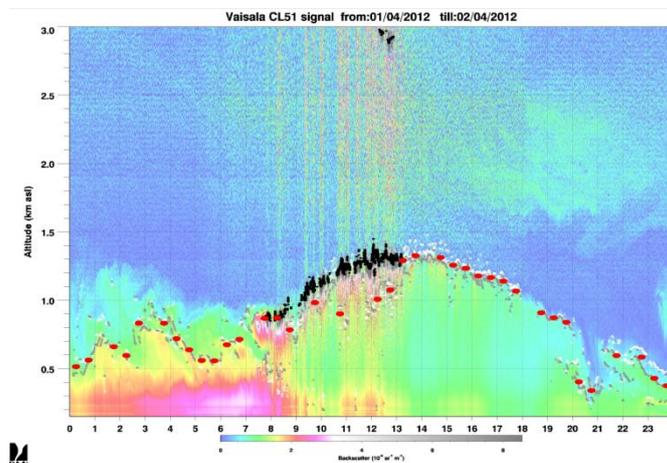


Figure 5: This figure shows the cloud base height (black dots) and the mixing layer height (red dots)

Task 3.2.3: Operate a mini-DOAS instrument at Ukkel

A mini-DOAS instrument has been continuously operating in Ukkel by BIRA-IASB RU2 since April 2011. In addition to the aerosol content (see Task 3.3 below), tropospheric NO₂ vertical column densities (VCDs) have been retrieved from mini-DOAS measurements and this data set has been used to validate GOME-2 satellite observations above Ukkel as illustrated in Figure 6.

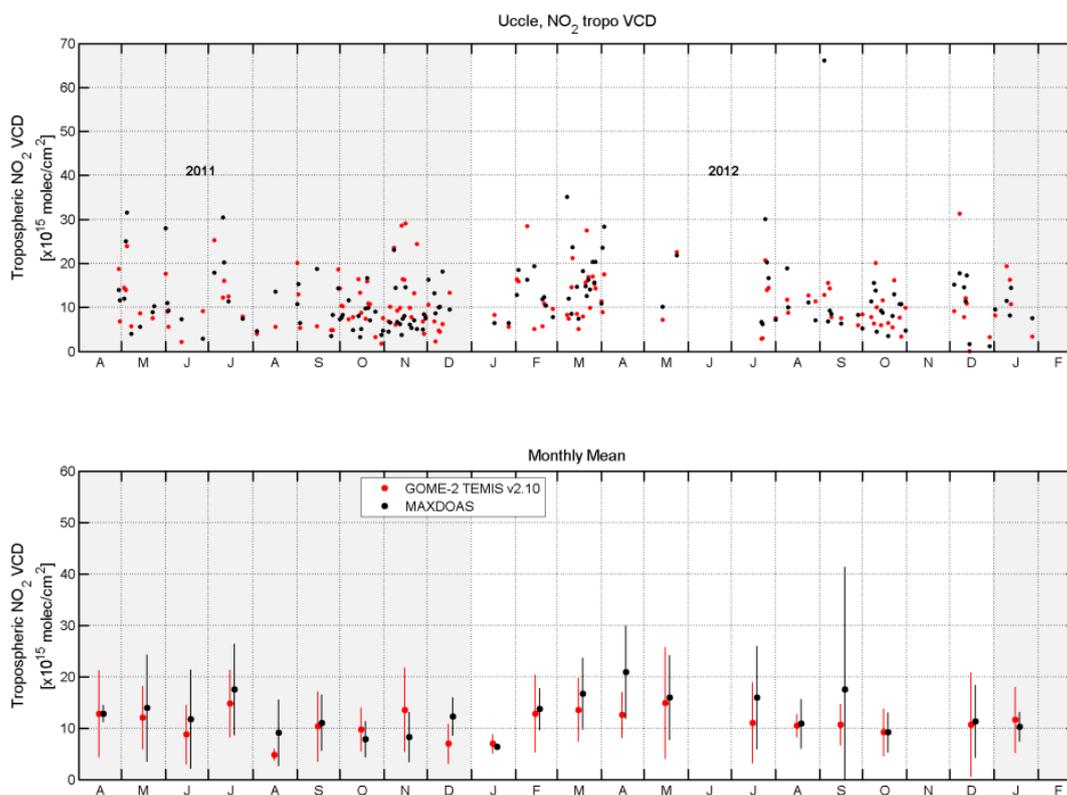


Figure 6: Time series of mini-DOAS and GOME-2 GDP4.5 tropospheric NO₂ VCD above Uccle since May 2011. The first panel presents the daily coincident points and the second panel presents the monthly mean values.

Generally, a good agreement is obtained between the mini-DOAS and the GOME-2 data, with similar capture of the pollution episodes and consistent seasonal variations, i.e. with high NO₂ in winter and low NO₂ in summer. However, larger differences between the monthly means are sometimes seen. This is likely related to the location of the mini-DOAS instrument, in the city center, where the measurements are more sensitive to local pollution peaks, while these tend to be attenuated in the satellite pixel.

Task 3.3 Interpretation of aerosol data at Ukkel

Task 3.3.1: Demonstrate the strength of the aerosol remote sensing by exploiting the ensemble of data and by deriving the maximum of information about the aerosol optical properties at Ukkel. RMI has different instruments with which the optical properties of aerosols can be determined. The measurements of the Brewer instruments can be used to retrieve AOD at 306, 310, 313, 316, 320 and 340nm (Cheymol & De Backer, 2003; De Bock, V. et al. 2010). Also preliminary SSA values are available at 306, 310, 313, 316 and 320nm. At the end of 2012, a nephelometer and an aethalometer have been purchased. Both instruments are not yet operational.

Figure 7 shows the comparison between retrieved AODs retrieved at 360 nm from mini-DOAS measurements at Ukkel and co-located CIMEL data (v1.5), for the year 2012. A reasonably good agreement is obtained with a correlation coefficient (R) of 0.69 and a slope (S) of 0.79. The correlation coefficient and slope values increase when removing high AOD events in both data sets (in this case, R=0.73 and S= 0.88). We are currently investigating the origin(s) of this feature. It could be related to the fact that the removal of high AOD events is acting as a cloud filter.

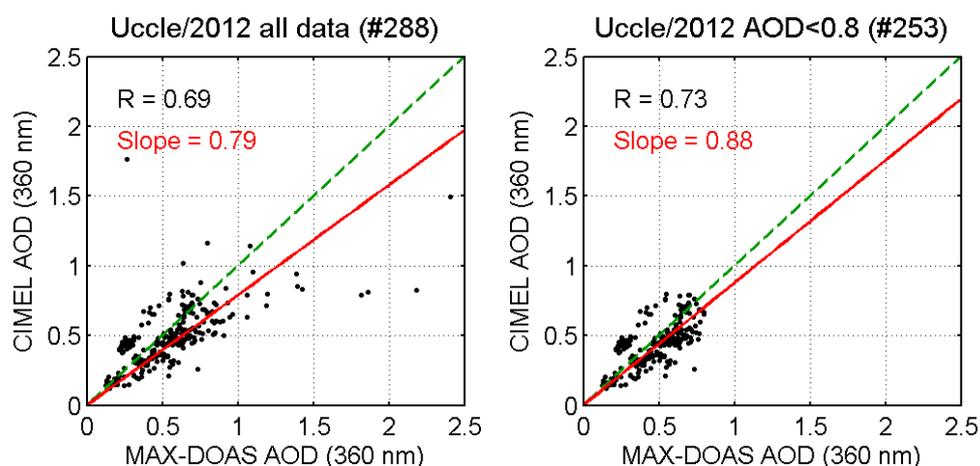


Figure 7: Scatter plots of the total AODs at 360 nm retrieved using O₄ MAX-DOAS measurements in Ukkel versus the values from co-located CIMEL data (v1.5).

Task 3.3.2: study the agreement between aerosol optical properties measured with the set of instruments mentioned above and simulated with Chimere

Chimere is now running and providing us with data going back to 1990. We have started the analysis of the format of the Chimere output data and have come to some conclusions on how to alter the output so that it can be used as input for OPAC. We need to change the unit of the Chimere output which is given in µg/m³ to particles/cm³. Also the aerosol classes are different in the two models. We now know how to redefine the output classes of Chimere into the input classes of OPAC.

Task 3.3.3: study the correlation between observed aerosol properties and source regions, using backtrajectories and meteorological parameters

We don't have results yet, due to the delay in the start of this task.

Task 3.3.4: Improve UV index forecast

Some adjustments and improvements have been made 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, which 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 2011.

WP4 – African emissions

We are ready to leave for Bujumbura for the installation of the MAXDOAS and CIMEL instruments. Also at the University of Bujumbura, the required construction for the housing of the instruments is ready. Eugène Ndenzako has returned to Burundi and will work with us locally during the installation and will be supporting the experiment once running.

WP 5 – Outreach

Task 5.1 To disseminate the results in the scientific community

Task 5.2.1: Via publications, workshops, international symposia

We have published several peer-reviewed papers and attended several international conferences and presented our work through oral and poster presentations. (See section 9 for a complete list of publications and communications).

Task 5.2.2: By submitting data to international databases

The Brewer ozone and UV data are deposited in the WOUDC database (www.woudc.org) and the Brewer AOD at 340nm are submitted to the GAW World Data Centre for aerosols, hosted by EBAS (<http://ebas.nilu.no/Default.aspx>).

NDACC data at Jungfraujoch and Reunion Island are submitted regularly to the NDACC databases (<ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/reunion/hdf/ftir>, and <ftp://ftp.cpc.ncep.noaa.gov/ndacc/station/jungfrau/hdf/ftir>). (At present, the VOC data at Reunion Island have not yet been submitted; this will happen soon.)

The TCCON data are available from the TCCON public archive (<http://tcon.ipac.caltech.edu/>).

The CIMEL sunphotometer are available from the AERONET Website. They have been used by other researchers, e.g., the H₂O total column data are used in the Solar-Terrestrial Center of Excellence, aerosol data have been solicited by Alina Sydorenko, researcher of the Ukrainian Hydrometeorological Institute, Kyiv, and they have been used in a publication by E. Nikitidou et al. (The aerosol forcing efficiency in the UV region and the estimation of single scattering albedo at a typical West European site, E. Nikitidou, A. Kazantzidis, V. De Bock and H. De Backer).

ULB spectroscopic data will be proposed for inclusion in HITRAN and GEISA

5. PRELIMINARY CONCLUSIONS AND RECOMMENDATIONS

Work Package 1: Direct Greenhouse gases

For the first time, approaches have been developed and successfully implemented for the retrieval of CCl_4 and CF_4 from ground-based FTIR spectra. In both cases, our investigations provided results in excellent agreement with findings from *in situ* surface monitoring networks. This is important for CCl_4 since unknown anthropogenic sources might be responsible for the discrepancies noted between emissions derived from the observations (40-80 Gg/yr) and emissions reported to UNEP (0-30 Gg/yr). For CF_4 , our results allow confirming the slowing down in its recent emissions, despite an AI production on the rise. Nevertheless, efforts by the AI industry should be pursued to further limit emissions of a very long-lived and potent GHG. Also, it should be pointed out that there is room for improvement of the CCl_4 and CF_4 spectroscopic line parameters which remain characterized by large uncertainties, affecting the systematic error budget and hence the determination of precise absolute concentrations for these GHGs from the remote-sensing technique.

At Jungfraujoch, consistent timeseries over the period 1983 to present of the major greenhouse gases CH_4 and N_2O have been built and first trends over this period have been derived. These two data sets combine in a consistent way column data from three successive instruments.

The AGACC-II project has enabled us to make Reunion Island become a qualified TCCON observatory, which is unique in the southern hemisphere subtropics. We have started the buildup of a timeseries of high-precision column-averaged dry-air mole fractions of the major greenhouse gases CO_2 , CH_4 , and N_2O . The results of the ongoing study about the consistency between the NDACC measurements of CH_4 in the mid-infrared and the newer TCCON measurements in the near-infrared will be very important to link both networks and to make use of both networks (with different spatial and temporal coverage) in ongoing and future satellite data validation (e.g., in the framework of the ESA Climate Change Initiative Greenhouse Gases project) and inverse model studies.

Work Package 2: Volatile Organic Compounds (VOC) and CFC-substitutes in the troposphere

As to methyl chloride, the recent release of spectroscopic parameters for CH_3Cl and C_2H_6 will hopefully help to solve the issues encountered during our first retrieval attempts, with preliminary time series showing a seasonal modulation out of phase and columns too large by about 50%. We have now to verify that the poor quality of the CH_4 line parameters (a strong interference) will not further hamper the – precise– retrieval of CH_3Cl .

Regarding the retrieval of methanol from the Jungfraujoch, a robust strategy has been developed, with information content well above our expectations. However, large systematic residuals suggest that the quality of the CH_3OH line parameters is subject to improvements.

The observations of the VOCs at Reunion Island combined with satellite data and model studies have shed light on the uncertainties in the fire emission databases and in the budgets of these species. In particular, our measurements have contributed to the finding that there is a significant missing source in the budget of formic acid. It has been found that the annual formic acid source is 2-3 times higher than estimated from known sources. This has important implications on precipitation acidity. These findings stimulate further studies searching for the missing source.

The tasks 2.2.1.a-c devoted to the improvement of reference spectroscopic information for H_2CO , C_2H_6 and C_2H_4 are progressing well. In particular, we were able to improve and extend line assignments for the ν_7 band of ethane, despite an unsuccessful initial analysis. This work and the task devoted to ethylene are on schedule. However, we faced two unexpected difficulties with the analysis of the IR spectra of formaldehyde, one being essentially solved and the other still not understood. These problems prevented us from providing deliverables D2.5 in due time (M18). As all the related tasks are ongoing, it is too early to provide recommendations for the reference spectroscopic information.

Work Package 3: Aerosol properties and radiative forcing at Ukkel

New instruments and tools have been implemented and developed for detection and characterisation of aerosol. It has been demonstrated that the Brewer direct sun measurements and sun scan measurements are suited for the retrieval of AOD values. The cloud screening procedure, which was developed during the first year of AGACC-II for the AOD retrieval using Brewer sun scan measurements, generates reliable Brewer AOD data and is now also applied to the AOD retrieval

using direct sun measurements. This cloud screening procedure could serve as inspiration for the cloud screening procedure of the MAXDOAS.

The preliminary algorithm which is currently used to determine SSA values (for cloudless days) gives unrealistically low values for days with low AOD. Also the range of SSA values is much larger on days with low AOD values. We need to look into the sensitivity of the model to the input parameters (such as ozone, AOD) but also to the accuracy of the measured UV irradiance in order to search for ways to improve the algorithm

Progress has also been made with aerosol retrievals from MAX-DOAS measurements – although with some delay due to personnel problems: a major effort will be devoted to this activity in the second half of the project.

Work package 4: African emissions

All preparatory work has been done to start the operation of an atmospheric observatory at the University of Bujumbura in Burundi early 2013. The contacts with the local researchers have been tightened and are very encouraging.

6. PROSPECTS AND FUTURE PLANNING

Work Package 1: Direct Greenhouse gases

The results for CF₄ above Jungfraujoch will be finalized and published in a peer-reviewed scientific journal.

The available CO₂ lines in the Jungfraujoch grating spectra will be identified and it will be checked that they can be used to derive robust total column results. If yes, the CO₂ time series will be extended back in time and trend analyses will be performed

The TCCON measurements at Reunion Island will be continued. An apparent seasonal variation of NO₂ will be investigated. The study concerning the consistency between NDACC and TCCON data for CH₄ will be finalised and the appropriate conclusions will be drawn.

Work Package 2: Volatile Organic Compounds (VOC) and CFC-substitutes in the troposphere

At Jungfraujoch, it is planned to

- Optimize the retrieval strategy for H₂CO from high altitude FTIR spectra, characterize the information content and start comparison with MAXDOAS measurements. The MAXDOAS analyses will be updated according to the lessons learned during the CINDI campaign.
- Implement the most recent line parameters for CH₃Cl, C₂H₆ and CH₄, re-evaluate the possibility to produce sensible results for CH₃Cl
- If CH₃Cl and HCFC-142b retrievals are successful, include their contributions in the CCl₄ budget and update the trend analyses; compare with AGAGE and ESRL-NOAA findings
- Perform first CF_y budget investigations from the FTIR technique, including the contributions of CFC-11, -12, HCFC-22, -142b, CF₄, evaluate its representativity and compare with AGAGE and NOAA-ESRL results.

At Reunion Island,

- More work will be done on the retrieval of PAN, acetone and CH₃Cl: some studies were already performed, in particular for PAN, and it is not sure that it will be feasible to retrieve these species at St. Denis. However, a new similar FTIR instrument will be installed in February 2013 at the Maito mountain site at Ile de La Réunion, at 2000 masl, and the feasibility checks will be repeated for this site; the smaller humidity at this site may enhance the chances to retrieve these species.
- If new spectroscopic data become available, the timeseries for C₂H₄, C₂H₆ and H₂CO will be updated.

The tasks 2.2.1.a-c devoted to the improvement of reference spectroscopic data for H₂CO, C₂H₆ and C₂H₄ will be completed. The work will be reported in articles to be published in peer-reviewed journals. Specifically:

- We will analyse the FIR and IR spectra recorded during the three remaining experiments, and try to resolve the inconsistencies observed for the TDL spectra recorded in the range 2928.4–2929.2 cm⁻¹. The absolute intensity of the H₂CO IR lines probed by the TDL spectrometer thus determined will be compared to HITRAN (Rothman et al, 2009) and used to “calibrate” the line intensities available therein. The quality of the modeling of the whole ν_1 and ν_5 band system (2500–3200 cm⁻¹) using the improved linelist (deliverable D2.5) will be assessed using FTS spectra recorded, with particular attention to the regions used to probe H₂CO in the terrestrial atmosphere (Vigouroux et al, 2009).
- We will generate a linelist for the ν_7 band of ethane [deliverable D2.4, in HITRAN format (Rothman et al, 2009)]. This information will be complemented with literature data for the lower state energies and air- and self-broadening and shift coefficients. The quality of this linelist will be assessed using laboratory spectra, and will then be sent to the ULg and BIRA partner for validation with atmospheric spectra.
- The frequency analysis and line intensity measurements in the 10 μ m region of the ethylene spectrum will be completed. The measured line intensities will be fit with the French model to determine parameters of the dipole moment function of ethylene. A linelist [deliverable D2.3, in HITRAN format (Rothman et al, 2009)] will be generated using this theoretical model and all the parameters thus determined, and compared with HITRAN (Rothman et al, 2009).

Work Package 3: Aerosol properties and radiative forcing at Ukkel

MAX-DOAS related activities within Work Package 3 have not progressed as expected due to the long-term absence of Dr. K. Clémer who was in charge of MAX-DOAS aerosols aspects within BIRA-IASB RU2 combined to the difficulty of hiring personnel. Since October 1st 2012, a new collaborator, Dr. C. Gielen, is working full time in BIRA-IASB RU2 on MAX-DOAS aerosols retrievals within AGACC-II. We hope to catch up with the delays in MAX-DOAS activities by the end of the project.

In the next part of the project, the aerosol extinction vertical profiles and corresponding AODs will be retrieved at Ukkel by applying the bePRO tool to the whole time-series of mini-DOAS measurements (April 2011 till now). In collaboration with KMI-IRM, the seasonal and diurnal variations of the aerosol content in Ukkel will be investigated using MAX-DOAS data as well as observations from collocated CIMEL, lidar ceilometer, nephelometer, and Brewer instruments. The possibility of retrieving aerosol optical properties like phase function and single scattering albedo (SSA) from MAX-DOAS measurements will be also further investigated.

We will also continue the routine operation of the Brewer instrument at Ukkel and will compare the Brewer measurements with those of the CIMEL on a regular basis.

The results obtained within task 3.1.2 during the first two years show that the SSA-algorithm applied to the Brewer data performs poorly in conditions with low AOD (<0.5). In the second phase of the project, we will look into the sensitivity of the algorithm to the input parameters of the TUV model and the accuracy of the measured UV irradiance in order to look for further improvements of the algorithm.

The results of our Brewer time series analysis (which includes a trend analysis, change point analysis and multiple regression analysis) of erythemal UV dose, total solar radiation, total ozone and AOD at 320nm will be published in 2013.

We will continue the routine operation of the lidar and we will also continue the comparison of the MLH retrieved by the lidar with the ones from ECMWF and ALARO. We are currently in the process of buying 3 new instruments which will be installed in different places in Belgium in 2013.

We will continue to monitor the optical properties of aerosols with as many instruments as possible. Next to the instruments that are already operational (Brewer, Lidar, Cimel), two new instruments will be installed, a nephelometer and an aethalometer, which will provide us with information on the scattering and absorption coefficients of particles. These two instruments could also be used for campaigns in Bujumbura.

After applying the changes mentioned in section 4 to the Chimere output format, we will continue task 3.3.2 as planned and use the modelled chemical composition of aerosols (provided by Chimere) to model the optical properties. These modelled properties (AOD and SSA) will then be compared to the measured ones in order to determine whether the combination of Chimere/OPAC is suited for the prediction of the optical properties of aerosols.

Backtrajectory modelling will be used to determine the source region of the aerosols above Ukkel in order to investigate whether there is a correlation between the source region of the aerosols (the clusters) and the observed AOD and SSA. The relation between AOD/SSA and meteorological conditions will be studied in detail. With these results, an attempt will be made to assign tendencies in AOD/SSA to either the origin of the air masses/meteorological conditions or to changes in emissions.

Currently the UV index forecast model uses a constant SSA value and climatological monthly means for the AOD. The forecast can be improved by using modelled SSA and AOD. We will examine which of the two methods (Chimere/OPAC modelling or backtrajectory analysis) is best suited for this.

We have also prepared tools to perform some direct radiative forcing calculations of aerosol, based on the ALVL code. ALVL is an advanced radiative transfer software package developed at BIRA-IASB in the past few years. So when the aerosol data will become available, evaluations of their direct radiative forcing will be performed.

The largest challenge in the next 2 years will be to exploit all the aerosol data together with the models in order to gain a maximum of information about their physical, chemical and radiative properties, and source regions.

Work package 4: African emissions

In spring 2013, a CIMEL and a MAXDOAS instrument will be installed at the University of Bujumbura and the data processing chain will be set up. Local support will be available thanks to the education of Eugène Ndenzako during his stay at BIRA from November 2012 to early February 2013. In the next 2 years (2013-2014) we will then start the exploitation of the MAXDOAS measurements (NO₂, glyoxal, H₂CO, and aerosol) in Bujumbura, including the interpretation using ancillary data from 3D chemistry transport models and satellite measurements, and later (2014) start the investigation of transport of emissions and their impact on measurements in Bujumbura and Reunion Island (using FLEXPART dispersion modeling).

Work package 5: Outreach

Some papers have already been published and new manuscripts are in preparation. We expect more publications to come in the next 2 years. Various participations to conferences and workshops are planned. In particular, we report about the AGACC-II activities at the annual TCCON meetings and NDACC Working Group and Steering Committee meetings.

We will continue to submit the data to the dedicated databases.

In 2013, RMI will start participating in a new COST action (COST ES1207: A European Brewer Network – EUBREWNET) which will coordinate Brewer spectrophotometer measurements of ozone, spectral UV and AOD in the UV within Europe.

Activities in support of decision making, like the ones mentioned in Section 10, will be continued and reinforced in the second half of the project, when more results become available.

7. FOLLOW-UP COMMITTEE

Some members of the Follow-Up Committee have attended our annual progress meetings and have participated actively to the discussions: their questions and advices are very useful for the project. Particular strong interactions exist with Dr. J.-F. Müller and his team at BIRA and with the team of Dr. P.-F. Coheur at ULB: our data are supporting the modelling and satellite validation studies.

The presence of Jan Voet in the Follow-Up Committee, who is the national contact point for Copernicus in-situ data, has made our activities and data better known to the Belgian Copernicus users group and to EEA who is coordinating the in-situ component of Copernicus.

Unfortunately, not all members of the Follow-Up Committee show up at our meetings, despite our repeated invitations and despite the fact that we selected them from the scientific communities in Belgium and neighbour countries that deal with related research.

The interaction with the Committee remains limited to an annual meeting.

8. REFERENCES

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9. PUBLICATIONS / VALORISATION

9.1 Publications of the teams

9.1.1 Peer review

BIRA-IASB

2011

Merlaud, A., M. Van Roozendael, N. Theys, C. Fayt, C. Hermans, B. Quennehen, A. Schwarzenboeck, G. Ancellet, M. Pommier, J. Pelon, J. Burkhart, A. Stohl, and M. De Mazière, Airborne DOAS measurements in Arctic: vertical distributions of aerosol extinction coefficient and NO₂ concentration, doi:10.5194/acp-11-9219-2011, Atmos. Chem. Phys., 11, 9219–9236, 2011. (See Annex, publication No. 1)

Senten, C., M. De Mazière, G. Vanhaelewyn, and C. Vigouroux, Information operator approach applied to the retrieval of the vertical distribution of atmospheric constituents from ground-based high-resolution FTIR measurements, Atm. Meas. Tech., 5, 161-180, 2012. (See Annex, publication No. 3)

Zieger, P., E. Weingartner, J. Henzing, M. Moerman, G. de Leeuw, J. Mikkila, K. Clémer, M. Van Roozendael, S. Yilmaz, U. Frieß, H. Irie, T. Wagner, R. Shaiganfar, S. Beirle, A. Apituley, K. Wilson, and U. Baltensperger, Comparison of ambient aerosol extinction coefficients obtained from in-situ, MAX-DOAS and LIDAR measurements, Atmos. Chem. Phys., 11, 2603-2624, 2011. (See Annex, publication No. 4)

2012

Hendrick, F., E. Mahieu, G. E. Bodeker, K. F. Boersma, M. P. Chipperfield, M. De Mazière, I. De Smedt, P. Demoulin, C. Fayt, C. Hermans, K. Kreher, B. Lejeune, G. Pinardi, C. Servais, R. Stübi, R. van der A, J.-P. Vernier, and M. Van Roozendael, Analysis of stratospheric NO₂ trends above Jungfraujoch using ground-based UV-visible, FTIR, and satellite nadir observations, Atmos. Chem. Phys., 12, 8851-8864, 2012. (See Annex, publication No. 5)

Merlaud, A., M. De Mazière, C. Hermans, and A. Cornet, Equations for solar tracking, Sensors, 12, 4047-4090, 2012. doi:10.3390/s120404074. (See Annex, publication No. 6)

Pinardi, G., Van Roozendael, M., Abuhassan, N., Adams, C., Cede, A., Clémer, K., Fayt, C., Frieß, U., Gil, M., Herman, J., Hermans, C., Hendrick, F., Irie, H., Merlaud, A., Navarro Comas, M., Peters, E., Piters, A. J. M., Puentedura, O., Richter, A., Schönhardt, A., Shaiganfar, R., Spinei, E., Strong, K., Takashima, H., Vrekoussis, M., Wagner, T., Wittrock, F., and Yilmaz, S.: MAX-DOAS formaldehyde slant column measurements during CINDI: intercomparison and analysis improvement, Atmos. Meas. Tech., 6, 167-185, doi:10.5194/amt-6-167-2013, 2013. (See Annex, publication No. 7)

Senten, C., M. De Mazière, G. Vanhaelewyn, and C. Vigouroux, Information operator approach applied to the retrieval of the vertical distribution of atmospheric constituents from ground-based high-resolution FTIR measurements, Atm. Meas. Tech., 5, 161-180, 2012. (See Annex, publication No. 8)

Vigouroux, C., T. Stavrou, C. Whaley, B. Dils, V. Duflot, C. Hermans, N. Kumps, J.-M. Metzger, F. Scolas, G. Vanhaelewyn, J.-F. Müller, D. B. A. Jones, Q. Li, and M. De Mazière, FTIR time-series of biomass burning products (HCN, C₂H₆, C₂H₂, CH₃OH and HCOOH) at Reunion Island (21°S, 55°E) and comparisons with model data, Atmos. Chem. Phys., 12, 10367-10385, 2012. (See Annex, publication No. 9)

ULB

2011

Lattanzi, F., C. di Lauro, J. Vander Auwera, « Toward the understanding of the high resolution infrared spectrum of C₂H₆ near 3.3 μm », Journal of Molecular Spectroscopy 267 (2011) 71-79. (See Annex, publication No. 10)

ULg 2012

Rinsland, C.P., E. Mahieu, P. Demoulin, R.Zander, C. Servais, and J.-M. Hartmann, Decrease of the Carbon Tetrachloride (CCl₄) Loading above Jungfraujoch, based on High Resolution Infrared Solar Spectra recorded between 1999 and 2011, *J. Quant. Spectrosc. Radiat. Transfer*, 113, 1322-1329, 10.1016/j.jqsrt.2012.02.016, 2012.[<http://hdl.handle.net/2268/121150>] (See Annex, publication No. 11)

9.1.2 Others

Theses

BIRA-IASB

PhD thesis of C. Senten (mid-2005 - 2012): ‘Development and evaluation of a modified OEM inversion algorithm for ground-based FTIR spectra. Application to spectra recorded at Réunion Island.’, promotor: Prof. F. Van Oystaeyen, co-promotor: M. De Mazière. PhD obtained in March 2012 at the University of Antwerp. (See Annex, publication No. 12)

ULg

- Master thesis:

Bader, W., Extension of the long-term total column time series of atmospheric methane above the Jungfraujoch station: analysis of grating infrared spectra between 1977 and 1989, master thesis, Université de Liège, 17 Allée du 6 Août, 4000-Liège, Belgium, pp. 1-95, 2011. [<http://hdl.handle.net/2268/99452>] (See Annex, publication No. 13)

- PhD thesis:

Duchatelet, P., Fluorine in the atmosphere: Inorganic fluorine budget and long-term trends based on FTIR measurements at Jungfraujoch, Thèse de Doctorat, Université de Liège, 17 Allée du 6 Août, 4000-Liège, Belgique, 182 pages, mai 2011. [<http://hdl.handle.net/2268/91413>] (See Annex, publication No. 14)

Communications

BIRA-IASB

De Mazière, M., and M. Van Roozendael, Spectrometry and atmospheric research: a fruitful marriage, invited lecture at the 25 Spektrometertagung (25th Conference of Spectrometry), Schaffhausen, 26-28/9/2011. . (See Annex, publication No. 15)

De Mazière, M., Site Report for Ile de la Réunion, oral presentation at the TCCON/IRWG Meeting, NCAR, Boulder, CO, May 23-25, 2011. . (See Annex, publication No. 16)

De Mazière, M., Site Report for Ile de la Réunion, oral presentation at the TCCON/IRWG Meeting, Wengen, Switzerland, June 11-15, 2012. . (See Annex, publication No. 17)

Desmet, F., M. De Mazière, C. Hermans, F. Scolas, N. Kumps, TCCON Site Report for Île de la Réunion, oral presentation at TCCON/IRWG Meeting, Wengen, Switzerland, June 11-15, 2012. (See Annex, publication No. 18)

Desmet, F., C. Hermans, M. De Mazière, N. Kumps, F. Scolas, J-L. Baray, J-M. Metzger, The First Results from the New TCCON Station at Reunion Island, poster presentation at the EGU General Assembly, Vienna, April 2012. (See Annex, publication No. 19)

Dils, B., J. Cui, S. Henne, E. Mahieu, M. Steinbacher, M. De Mazière, 1997-2007 CO at the high Alpine site Jungfraujoch: A comparison between NDIR surface in situ and FTIR remote sensing observations, Seminar at BIRA, Brussels, 24 June 2011. (See Annex, publication No. 20)

Hendrick, F., P. Demoulin, K. Kreher, M. De Mazière, C. Fayt, C. Hermans, B. Lejeune, C. Servais, and M. Van Roozendael, Trend analysis of stratospheric NO₂ above Jungfraujoch (46.5°E, 8.0°E) using long-term ground-based UV-visible and FTIR observations, oral presentation at the European Geophysical Union General Assembly 2011, Vienna, Austria, 03–08 April 2011. (See Annex, publication No. 21)

- Hendrick, F., E. Mahieu, A. Rozanov, K. F. Boersma, M. De Mazière, P. Demoulin, C. Fayt, C. Hermans, G. Pinardi, and M. Van Roozendael, Trend analysis of stratospheric NO₂ above Jungfraujoch (46.5°N, 8°E) and Harestua (60°N, 11°E) using long-term ground-based UV-visible, FTIR, and satellite observations, poster presentation at the ESA Atmospheric Science Conference 2012, Bruges, Belgium, 18-22 June 2012. (See Annex, publication No. 22)
- Hendrick, F., D. Ionov, F. Goutail, A. Pazmino, U. Friess, M. Gil, J.-C. Lambert, M. Navarro, M. Pastel, J.-P. Pommereau, A. Richter, T. Wagner, F. Wittrock, and M. Van Roozendael, New NDACC recommendations for the retrieval of stratospheric NO₂ from ground-based zenith-sky UV-visible observations, Poster presentation at the General Assembly of the European Geosciences Union, Vienna, Austria, 22-27 April 2012. (See Annex, publication No. 23)
- Pinardi, G., C. Adams, S. Beirle, A. Cede, U. Friess, M. Gil, F. Hendrick, H. Irie, E. Peters, A. Piders, O. Puentedura, A. Richter, R. Shaigan, E. Spinei, K. Strong, H. Takashima, T. Wagner, F. Wittrock, S. Yilmaz, and M. Van Roozendael, Intercomparison of MAXDOAS HCHO slant columns during the CINDI campaign, poster presentation at the EGU General Assembly 2012, Vienna, Austria, 22-27 April 2012. (See Annex, publication No. 24)
- Pinardi, G., I. Van Roozendael M., J.-C. Lambert, K. Clemer, I. De Smedt, F. Hendrick, C. Lerot, N. Theys, J. van Gent, T. Vlemmix, M. De Mazière, H. De Backer, A. Decloo, H. Yu, Integrated Trace Gas Validation and Quality Assessment System for the EUMETSAT Polar System, oral presentation at the 2012 EUMETSAT Meteorological Satellite Conference, Sopot, Poland, 3-7 September 2012. (See Annex, publication No. 25)
- Pommereau, J.-P., F. Goutail, A. Pazmino, D. Ionov, F. Hendrick, and M. Van Roozendael, Evaluation of Satellites Total Ozone and NO₂ columns retrievals using the NDACC-SAOZ UV-Vis Network, poster presentation at the 39th COSPAR Scientific Assembly, 14-22 July 2012. (See Annex, publication No. 26)
- Pommereau, J.-P., F. Goutail, A. Pazmino, D. Ionov, F. Hendrick, and M. Van Roozendael, Evaluation of Satellite Total Ozone and NO₂ Columns using the SAOZ UV-Vis Network, oral presentation at the ESA ATMOS 2012, Advances in Atmospheric Science and Applications, Bruges, Belgium, 18-22 June 2012. (See Annex, publication No. 27)
- Senten, C., M. De Mazière, G. Vanhaelewyn, and C. Vigouroux, Information operator approach applied to ground-based FTIR measurements at Ile de La Réunion and compared with optimal estimation approach, oral presentation at the NDACC Infrared Working Group Meeting, (NCAR, Boulder, CO, May 23-25, 2011), 2011. . (See Annex, publication No. 28)
- Valks, P., N. Hao, D. Loyola, I. De Smedt, M. Van Roozendael, J.-C Lambert, G. Pinardi, A. Delcloo and W. Zimmer, Tropospheric Trace-Gas Column Observations from GOME-2/MetOp, presentation at ESA atmospheric satellite conference, Bruges, 18-22 June 2012. (See Annex, publication No. 29)
- Vanhaelewyn, G., C. Vigouroux, M. De Mazière, Systematic error due to the choice in a priori profile, oral presentation by M. De Mazière at the NDACC Infrared Working Group Meeting, (NCAR, Boulder, CO, May 23-25, 2011), 2011. . (See Annex, publication No. 30)
- Vigouroux, C., T. Stavrou, C. Whaley, B. Dils, V. DufLOT, C. Hermans, N. Kumps, J.-M. Metzger, F. Scolas, G. Vanhaelewyn, J.-F. Müller, D. B. A. Jones, and M. De Mazière: FTIR time-series of biomass burning products (HCN, C₂H₆, C₂H₂, CH₃OH, and HCOOH) at Reunion Island and comparisons with model data, oral presentation at the IRWG/TCCON annual meeting, Wengen, 11-15 June, 2012. (See Annex, publication No. 31)
- Vigouroux C., T. Stavrou, C. Whaley, B. Dils, V. DufLOT, C. Hermans, N. Kumps, J.-M. Metzger, F. Scolas, G. Vanhaelewyn, J.-F. Müller, D. B. A. Jones, Q. Li, and M. De Mazière: Time-series of biomass burning products from ground-based measurements at Reunion Island and comparisons with model data, oral presentation at NDACC symposium, Saint-Paul, 7-10th November 2011. . (See Annex, publication No. 32)

KMI-IRM

- De Bock, V., H. De Backer and A. Mangold, Improved cloud screening for Aerosol Optical Depth measurements with a Brewer spectrophotometer, European Aerosol Conference, Manchester, United Kingdom, September 4-9, 2011. (See Annex, publication No. 33)
- De Bock, V., Aerosol optical depth measurements at 340 nm with a Brewer spectrophotometer and comparison with Cimel sunphotometer observations at Ukkel, Belgium, presentation at KMI-IRM, 5 October 2011. (See Annex, publication No. 34)

- De Bock, V., H. De Backer, A. Mangold, "Retrieval of Single Scattering Albedo values from Brewer spectrophotometer irradiance measurements at Uccle, Belgium", European Aerosol Conference 2012, Granada, Spain, 2-7 September 2012. (See Annex, publication No. 35)
- De Bock, V., H. De Backer, R. Van Malderen, "Analysis of an extensive time series of UV irradiation and AOD measurements in the UV-B region at Uccle, Belgium", International Radiation Symposium 2012, Berlin, Germany, 6-10 August 2012. (See Annex, publication No. 36)
- De Bock, V., H. De Backer, R. Van Malderen, "Analysis of an extensive time series of UV irradiation and AOD measurements in the UV-B region at Uccle, Belgium", Quadrennial Ozone Symposium 2012, Toronto, Canada, 26-31 August 2012. (See Annex, publication No. 37)
- Nikitidou, E., V. De Bock, H. De Backer and A. Kazantzidis, Estimation of aerosol optical properties and their effect on UV irradiance at Ukkel, Belgium, 11th EMS Annual Meeting / 10th European Conference on Applications of Meteorology (ECAM), Berlin, Germany, September 12-16, 2011. (See Annex, publication No. 38)
- Nikitidou, E., V. De Bock, H. De Backer, A. Kazantzidis, Aerosols optical properties and their effect on the UV solar irradiance at Uccle, Belgium, International Radiation Symposium 2012, Berlin, Germany, 6-10 August 2012. (See Annex, publication No. 39)

ULB

- Bourgeois, M.-T., M. Rotger, V. Boudon, J. Vander Auwera, « Frequency analysis of the 10 and 3 μm regions of the ethylene spectrum using the D_{2h} Top Data System », poster presentation at
- Colloque commun de la Division de Physique Atomique, Moléculaire et Optique de la Société française de Physique et des Journées de Spectroscopie Moléculaire, Metz, France, 3-6 July 2012 ;
 - 11th ASA Conference united with the 12th HITRAN Conference, Reims, France, 29-31 August 2012 ;
 - 22nd International Conference on High Resolution Molecular Spectroscopy, Prague, Czech Republic, 4-8 September 2012.
- Fissiaux, L., T. Földes, F. Kwabia Tchana, L. Daumont, M. Lepère, J. Vander Auwera, « Infrared line intensities for formaldehyde from simultaneous measurements in the infrared and far infrared spectra ranges », oral presentation (MG5), 66th Ohio State University International Symposium on Molecular Spectroscopy, Columbus, Ohio, USA, 20-24 June 2011.
- Fissiaux, L., T. Földes, F. Kwabia Tchana, L. Daumont, M. Lepère, J. Vander Auwera, « Infrared line intensities for formaldehyde from simultaneous measurements in the infrared and far infrared spectra ranges », poster presentation (O13), 22nd Colloquium on High Resolution Molecular Spectroscopy, Université de Bourgogne, Dijon, France, 29 August – 2 September 2011 (<http://agacc.aeronomie.be/>). (See Annex, publication No. 23)
- Tudorie, M., C. di Lauro, F. Lattanzi, J. Vander Auwera, « A new analysis of the ν_7 band of ethane », poster presentation at the Solvay workshop on femto-, astro-, spectro-ethyne, Université Libre de Bruxelles, Brussels, 2-5 May 2012.
- Tudorie, M., C. di Lauro, F. Lattanzi, J. Vander Auwera, « A new analysis of the ν_7 band of ethane », poster presentation at
- Colloque commun de la Division de Physique Atomique, Moléculaire et Optique de la Société française de Physique et des Journées de Spectroscopie Moléculaire, Metz, France, 3-6 July 2012 ;
 - 11th ASA Conference united with the 12th HITRAN Conference, Reims, France, 29-31 August 2012 ;
 - 22nd International Conference on High Resolution Molecular Spectroscopy, Prague, Czech Republic, 4-8 September 2012.
- Tudorie, M., C. di Lauro, F. Lattanzi, J. Vander Auwera, « An improved rotational analysis of the ν_7 band of ethane », oral presentation, International workshop on the spectroscopy of methane and derived molecules for atmospheric and planetary applications, Dole, France, 26-28 November 2012.

ULg

- Bader, W., B. Lejeune, P. Demoulin, P. Duchatelet, G. Roland, K. Sudo, H. Yashiro, and E. Mahieu, Extension of the long-term total column time series of atmospheric methane above the Jungfraujoch station: analysis of grating infrared spectra between 1976 and 1989, poster presentation at the “EGU 2011 General Assembly”, 3-8 April 2011, Vienna, Austria, 2011. (See Annex, publication No. 40)
- Bader, W., E. Mahieu, B. Bovy, B. Lejeune, P. Demoulin, C. Servais, First retrievals of methanol (CH₃OH) above Jungfraujoch (46.5°N): Optimization of retrieval strategy and information content, poster presentation at the “Journée des doctorants UNITER”, 10 December 2012, Bruxelles, Belgium, 2012. [<http://hdl.handle.net/2268/135878>] (See Annex, publication No. 41)
- Bader, W., A. Perrin, D. Jacquemart, J.J. Harrison, G.C. Toon, K. Sudo, O.A. Søvde, P. Demoulin, C. Servais, and E. Mahieu, Retrievals of ethane from ground-based high-resolution FTIR solar observations with updated line parameters: determination of the optimum strategy for the Jungfraujoch station, poster presentation at the “11th Atmospheric Spectroscopy Applications” meeting (ASA2012), united with the “12th HITRAN Conference”, 29 – 31 August 2012, Reims, France, 2012. [<http://hdl.handle.net/2268/129289>] (See Annex, publication No. 42)
- Bader, W., A. Perrin, D. Jacquemart, K. Sudo, H. Yashiro, M. Gauss, P. Demoulin, C. Servais, and E. Mahieu, Retrievals of ethane from ground-based high-resolution FTIR solar observations with updated line parameters: determination of the optimum strategy for the Jungfraujoch station, poster presentation at the “EGU 2012 General Assembly”, 22-27 April 2012, Vienna, Austria, 2012. [<http://hdl.handle.net/2268/117067>] (See Annex, publication No. 43)
- Bader, W., A. Perrin, D. Jacquemart, K. Sudo, H. Yashiro, O. A. Søvde, P. Demoulin, C. Servais, and E. Mahieu, Retrievals of ethane from ground-based high-resolution FTIR solar observations with updated line parameters: determination of the optimum strategy for the Jungfraujoch station, poster presentation at the “NDACC-IRWG/TCCON Meeting, 11-15, June 2012, Wengen, Switzerland, 2012. [<http://hdl.handle.net/2268/124230>] (See Annex, publication No. 44)
- Buschmann, M., S. Dohe, E. Mahieu, N. Deutscher, T. Warneke, and J. Notholt, CO₂ total column retrieval by mid-IR FT Spectroscopy, poster presentation at the “EGU 2012 General Assembly”, 22-27 April 2012, Vienna, Austria, 2012. [<http://hdl.handle.net/2268/118439>] (See Annex, publication No. 45)
- Demoulin, P., G. Roland, W. Bader, B. Lejeune, P. Duchatelet, E. Mahieu, C. Servais, and R. Zander, Analysis of historical grating spectra: Jungfraujoch atmospheric database extended back to 1977, oral presentation at the 2011 NDACC Symposium, 7-10 November 2011, Saint Paul, La Réunion, France, 2011. (See Annex, publication No. 46)
- Duchatelet, P., R. Zander, E. Mahieu, J. Mühle, P. Demoulin, B. Lejeune, G. Roland, C. Servais, and O. Flock, First retrievals of carbon tetrafluoride (CF₄) from ground-based FTIR measurements: production and analysis of the two-decadal time series above the Jungfraujoch, poster presentation at the “EGU 2011 General Assembly”, 3-8 April 2011, Vienna, Austria, 2011. (See Annex, publication No. 47)
- Mahieu, E., P. Duchatelet, R. Zander, B. Lejeune, W. Bader, P. Demoulin, G. Roland, C. Servais, C.P. Rinsland, M.J. Kurylo, and G.O. Braathen, Changes in atmospheric composition discerned from long-term NDACC measurements: trends in direct greenhouse gases derived from infrared solar absorption spectra recorded at the Jungfraujoch station, poster presentation at the World Climate Research Programme Open Science Conference, 24-28 October 2011, Denver, CO, USA, 2011. (See Annex, publication No. 48)
- Mahieu, E., P. Duchatelet, R. Zander, B. Lejeune, W. Bader, P. Demoulin, C. Servais, M. Schneider, S. Barthlott, and C.P. Rinsland, Long-term trends of a dozen direct greenhouse gases derived from infrared solar absorption spectra recorded at the Jungfraujoch station, poster presentation at the 2011 NDACC Symposium, 7-10 November 2011, Saint Paul, La Réunion, France, 2011. (See Annex, publication No. 49)
- Plass-Dülmer, C., S. Reimann, M. Wallasch, S. Solberg, D. Klemp, P. Coddeville, and E. Mahieu, NMHC Climatology from Central European Mountain Observatories, poster presentation at the “EGU 2011 General Assembly”, 3-8 April 2011, Vienna, Austria, 2011. (See Annex, publication No. 50)

9.2 Co-publications

9.2.1. Peer review

BIRA-ULB

2011

Stavrakou, T., A. Guenther, A. Razavi, L. Clarisse, C. Clerbaux, P.-F. Coheur, D. Hurtmans, F. Karagulian, M. De Mazière, C. Vigouroux, C. Amelynck, N. Schoon, Q. Laffineur, B. Heinesch, M. Aubinet, C. Rinsland, and J.-F. Müller, First space-based derivation of the global methanol emission fluxes, *Atmos. Chem. Phys.*, 11, 4873–4898, 2011. (See Annex, publication No. 51)

2012

Stavrakou, T., J.-F. Muller, J. Peeters, A. Razavi, L. Clarisse, C. Clerbaux, P.-F. Coheur, D. Hurtmans, M. De Mazière, C. Vigouroux, N. Deutscher, D. Griffith, N. Jones, C. Paton-Walsh, Satellite Evidence for a Large Source of Formic Acid From Boreal and Tropical Forests, *Nature Geosciences*, doi:10.1038/ngeo1354, 5, 26-30, 2012. (See Annex, publication No. 52)

BIRA-ULg

2011

Angelbratt, J., J. Mellqvist, D. Simpson, J. E. Jonson, T. Blumenstock, T. Borsdorff, P. Duchatelet, F. Forster, F. Hase, E. Mahieu, M. De Mazière, J. Notholt, A. K. Petersen, U. Raffalski, C. Servais, R. Sussmann, T. Warneke, and C. Vigouroux, Carbon monoxide (CO) and ethane (C₂H₆) trends from ground-based solar FTIR measurements at six European stations, comparison and sensitivity analysis with the EMEP model, doi:10.5194/acp-11-9253-2011, *Atmos. Chem. Phys.*, 11, 9253–9269, 2011. (See Annex, publication No. 53)

Dils, B., J. Cui, S. Henne, E. Mahieu, M. Steinbacher, M. De Mazière, 1997-2007 CO trend at the high Alpine site Jungfraujoch: A comparison between NDIR surface in situ and FTIR remote sensing observations, doi:10.5194/acp-11-6735-2011, *Atmos. Chem. Phys.*, 11, 6735–6748, 2011. (See Annex, publication No. 54)

2012

Kohlhepp, R., Ruhnke, R., M.P. Chipperfield, M. De Mazière, J. Notholt, S. Barthlott, R.L. Batchelor, R.D. Blatherwick, Th. Blumenstock, M.T. Coffey, P. Duchatelet, H. Fast, W. Feng, A. Goldman, D.W.T. Griffith, K. Hamann, J.W. Hannigan, F. Hase, N.B. Jones, A. Kagawa, Y. Kasai, O. Kirner, R. Kohlhepp, W. Kouker, I. Kramer, R. Lindenmaier, E. Mahieu, R.L. Mittermeier, B. Monge-Sanz, I. Murata, H. Nakajima, I. Morino, M. Palm, C. Paton-Walsh, Th. Reddmann, M. Rettinger, C.P. Rinsland, E. Rozanov, M. Schneider, C. Senten, B.-M. Sinnhuber, D. Smale, K. Strong, R. Sussmann, J.R. Taylor, G. Vanhaelewyn, T. Warneke, C. Whaley, M. Wiehle, and S.W. Wood, Senten, C., B.-M. Sinnhuber, D. Smale, K. Strong, R. Sussmann, J.R. Taylor, G. Vanhaelewyn, T. Warneke, C. Whaley, M. Wiehle, and S.W. Wood, Observed and simulated time evolution of HCl, ClONO₂, and HF total columns, *Atmos. Chem. Phys.*, 12, 3527-3556, 2012. (See Annex, publication No. 55)

BIRA-ULB-ULg

Duflot, V., Hurtmans, D., Clarisse, L., R'honi, Y., Vigouroux, C., De Mazière, M., Mahieu, E., Servais, C., Clerbaux, C., and Coheur, P.-F.: Measurements of hydrogen cyanide (HCN) and acetylene (C₂H₂) from the Infrared Atmospheric Sounding Interferometer (IASI), *Atmos. Meas. Tech. Discuss.*, 5, 7567-7586, (doi:10.5194/amtd-5-7567-2012), 2012. (See Annex, publication No. 56)

9.2.2 Others

BIRA-KMI-IRM

Pinardi G., M. Van Roozendaal, J.C. Lambert, K. Clemer, I. De Smedt, F. Hendrick, C. Lerot, N. Theys, J. van Gent, T. Vlemmix, M. De Mazière, H. Yu, H. De Backer, A. Delcloo, Integrated trace gas validation and quality assessment system for the EUMETSAT polar system, oral presentation at the 2012 EUMETSAT Meteorological Satellite Conference, Sopot, Poland, 3-7 September 2012. (See Annex, publication No. 57)

BIRA-ULg

- Dils, B., J. Cui, S. Henne, E. Mahieu, M. Steinbacher, M. De Mazière, NDIR surface in situ and FTIR remote sensing measurements at the Jungfraujoch see different CO trends, oral presentation at the EGU General Assembly 2011, Vienna, Austria, 6-8 April 2011. (See Annex, publication No. 58)
- Mahieu, E., J. Harrison, P.F. Bernath, G.C. Toon, C.P. Rinsland, P. Demoulin, P. Duchatelet, B. Lejeune, C. Servais, and M. De Mazière, First retrievals of methyl chloride from ground-based high-resolution FTIR solar observations, poster presentation at the “EGU 2011 General Assembly”, 3-8 April 2011, Vienna, Austria, 2011. (See Annex, publication No. 59)
- Mahieu, E., W. Bader, B. Bovy, B. Lejeune, C. Vigouroux, P. Demoulin, C. Servais, G. Roland, and R. Zander, Retrieval of methanol (CH₃OH) from the high-altitude Jungfraujoch station (46.5°N): preliminary total column time series, long-term trend and seasonal modulation, poster presentation at the “NDACC-IRWG Annual Meeting”, 11-15 June 2012, Wengen, Switzerland, 2012. [<http://hdl.handle.net/2268/124069>] (See Annex, publication No. 60)
- Mahieu, E., W. Bader, B. Lejeune, C. Vigouroux, P. Demoulin, C. Servais, G. Roland, and R. Zander, Seeking for the optimum retrieval strategy of methanol (CH₃OH) using ground-based high-resolution FTIR solar observations recorded at the high-altitude Jungfraujoch station (46.5°N), poster presentation at the “EGU 2012 General Assembly”, 22-27 April 2012, Vienna, Austria, 2012. [<http://hdl.handle.net/2268/116957>] (See Annex, publication No. 61)
- Vigouroux, C., M. De Mazière, P. Demoulin, C. Servais, T. Blumenstock, M. Schneider, F. Hase, R. Kohlhepp, S. Barthlott, J. Klyft, J. Mellqvist, M. Palm, J. Notholt, J. Hannigan, M. Coffey, R. Batchelor, Ozone tropospheric and stratospheric trends (1995-2011) at six ground-based FTIR stations (28°N to 79°N), poster presentation at the 2011 NDACC Symposium (St. Paul, La Réunion, Nov. 7-10, 2011). (See Annex, publication No. 62)
- Vigouroux, D., De Mazière, M., Demoulin, P., Servais, C., Hase, F., Blumenstock, T., Schneider, M., Regina Kohlhepp, Sabine Barthlott, Klyft, J., Personn, G., Palm, M., Notholt, J., J. Hannigan, M. Coffey: Ozone tropospheric and stratospheric trends (1995-2011) at six ground-based FTIR stations (28°N to 79°N), poster presentation at the Quadriennial Ozone Symposium QOS, Toronto, 27-31 August 2012. (See Annex, publication No. 63)

9.3 Other activities

ULG

We performed regular archiving of CH₄ and N₂O (as well as of non-AGACC-II targets) data at the NDACC host facility, using the new GEOMS hdf template. In 2012, we started to upload C₂H₆ archives to the same database.

We also organized and hosted the 2012 NDACC-IRWG meeting in Wengen, Switzerland (June 2012).

10. SUPPORT TO DECISION MAKING

ULg updated time series and trend values for the evolution of chlorine in the atmosphere for inclusion in the Tableau de Bord de l'Environnement Wallon and the MIRA-T report from the Vlaamse Milieu Maatschappij.

We promote the AGACC-II monitoring data in the GMES¹ in-situ Coordination activity (GISC), led by EEA.

The data are also reported in the annual GCOS report.

BIRA is valorising its TCCON activities in 2 EU FP7 projects, namely the InGOS project and the ICOS_Inwire project.

InGOS (Integrated non-CO₂ Greenhouse gas Observing System; <http://www.ingos-infrastructure.eu/>) is an EU funded IA (Integrating Activity) project targeted at improving and extending the European observation capacity for non-CO₂ greenhouse gases. The project will run from October 2011 to September 2015.

The ICOS-INWIRE project (ICOS Improved sensors, NetWork and Interoperability for GMES; <http://www.icos-inwire.lsce.ipsl.fr/>) started on 01 January 2013 and will finish in the end of year 2015.

The fundamental objective of the project is to enhance capabilities of the ICOS infrastructure for greenhouse gas monitoring, in order to meet the needs of operational users in the GMES Atmosphere and Land Core Service Elements. The GMES Atmosphere Service (GAS) aims at supporting the needs of policy makers.

In both projects, we aim at the integration of TCCON data in the ICOS infrastructure.

In particular in ICOS_Inwire, we have the commitment to deliver the Reunion TCCON data within 1 month after data acquisition. As such, they will be available to GAS and serve the needs of the GAS users, among which the policy makers.

The coordinator of AGACC-II is also coordinator of the EU FP7 project NORS (Demonstration Network Of ground-based Remote Sensing Observations in support of the GMES Atmospheric Service; <http://nors.aeronomie.be>), which aims at supporting the quality assessment and improvement of the Copernicus Atmosphere Service by the use of NDACC data. The MAXDOAS aerosol, HCHO and NO₂ data are included in the target products of NORS; so are CO and CH₄ from the FTIR spectrometers. All improvements to these products made in AGACC-II will therefore be beneficial to GAS and as such to policy makers and the public. In the longer term, more NDACC products will be incorporated in NORS-type efforts.

¹ GMES has been renamed Copernicus in December 2012

ANNEXES

A.1. Copy of the publications

A.2. Minutes of the follow-up committee meetings

A.3. Complementary information (survey form, data, calculation, etc.)

(annex A.3 is available for the experts but is not a formal part of the report).