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

«AGACC»

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SCIENCE FOR A SUSTAINABLE DEVELOPMENT (SSD)

Atmosphere

FINAL REPORT PHASE 1

SUMMARY

ADVANCED EXPLOITATION OF GROUND-BASED MEASUREMENTS FOR ATMOSPHERIC CHEMISTRY AND CLIMATE APPLICATIONS
«AGACC»

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The AGACC project (Advanced exploitation of Ground-based measurements for Atmospheric Chemistry and Climate applications) joins international scientific research regarding important environmental issues, that are our changing climate and degrading air quality. There is evidence for a strong link between the chemical composition of the atmosphere – including the troposphere and the stratosphere - and climate. Air quality is dominated by the boundary layer and lower troposphere composition, and the underlying emissions. AGACC therefore exploits past and current ground-based observations at Ukkel (50.5°N, 4°E), Jungfraujoch (ISSJ, 46.5°N, 8°E) and Ile de La Réunion (21°S, 55°E) for investigating chemical changes in the atmosphere that have an impact on the Earth's climate and/or air quality. In particular, it targets the detection and quantification of a number of atmospheric constituents that hitherto have not been looked at in depth, because they require advanced detection methods.

Various such advanced detection techniques are mastered in the AGACC consortium. Moreover, a good synergy between the various observation methods exists and is exploited in the project, as will be demonstrated in this report. It must also be mentioned that AGACC benefits from the international community in which the partners are involved – in particular the Network for the Detection of Atmospheric Composition Change (NDACC) and the COST Action WAVACS (Atmospheric Water Vapour and the Climate System; Action ES0604) that started in 2007.

The project is structured according to a number of activities and workpackages: the particular objectives of these are given hereafter, together with the associated results obtained thus far in the first phase of the project (2006-April 2008).

WP1000 Atmospheric Water Vapour

Water vapour is the most important greenhouse gas. There are indications that the amount of water vapour is changing. More investigations are needed to elucidate these changes and their origin. WAVACS is an example of the attention that is currently given on the international scene to this constituent. One of the major challenges is the measurement of water vapour in the upper troposphere / lower stratosphere, where large vertical gradients exist. Another complication is the fact that the abundance of water vapour is highly variable, in time and space. The determination of the water vapour isotopologues can shed light on the processes that underly its distribution in the atmosphere.

Remote sensing using Fourier transform infrared (FTIR) spectrometry is a candidate method to determine these isotopologues. Retrieval strategies for determining the amount and vertical distribution of the atmospheric water vapour above Uccle, Jungfraujoch and Ile de La Reunion from FTIR observations have been developed. Promising results have been obtained at all sites, for the primary as well as for some secondary isotopologues ($H_2{}^{16}O$, $H_2{}^{18}O$, HDO). Comparisons with the IASI satellite data, although very preliminary, demonstrated the complementarity of both datasets and the need for joint retrievals. The accurate detection of water vapour by FTIR must be pursued because the FTIR technique has the potential to record isotopologues amounts and isotopic ratios on a continuous basis, and therefore to provide valuable information about transport and dynamical processes. At Ukkel, profiles (up to about 8 to 12 km altitude) and integrated water vapour amounts have been compared with several other instruments (radiosoundings, CIMEL data, GPS measurements). The observed biases are always smaller than 20%. The investigation of the discrepancies will be pursued, and optimal retrieval strategies will be defined.

At Ukkel, a timeseries of radiosoundings exists going back to 1990. It is known that such measurements are indebted with a number of artefacts. The Leiterer method (2005) has been found to be the better method to correct for these artefacts. After its application to the Uccle soundings, preliminary trends for the relative humidity in the upper troposphere and at the tropopause have been determined. We observe an increasing humidity before 2001 at a rate of 0.97%/year and a strong decrease since then, by -3.57%/year. These changes seem to be caused by a lifting, respectively lowering of the tropopause The causes for these changes are not yet understood. .

Further work will include the investigation of temporal and spatial variabilities of the atmospheric water vapour abundance.

WP 1200 Aerosol

The CIMEL instrument was installed at Uccle (50.5°N, 4°E) in summer 2006. The sunphotometer operated correctly from July 2006 until September 2007. During the time period October 2007-Februari 2008 the instrument was recalibrated at Carpentras (44° N, 5° E). Since March 2008 the recalibrated sunphotometer is fully operational again. The processed data, e.g., aerosol optical depth (AOD), aerosol size distribution, and integrated water vapor (IWV), are available on the AERONET website (http://aeronet.gsfc.nasa.gov/), and have been used in several studies since then, among which a comparison with aerosol data derived from the Brewer instrument at Uccle.

A new MAXDOAS instrument has been built, with a configuration that is optimised for enabling the determination of aerosol optical depth and optical properties. It allows the collection of scattered solar light, as well as direct sun measurements using a sun tracker. First tests of the instrument at Uccle are ongoing. A first version of an aerosol inversion algorithm for the MAXDOAS spectra has been developed. It has the capability to retrieve aerosol extinction vertical profiles and total aerosol optical depths based on measurements of the O_4 slant column densities (using the DOAS technique) and relative changes in intensities. An important improvement of this algorithm compared to previously reported retrieval algorithms, is the implementation of the radiative transfer code LIDORT in the forward model. This enables an analytical calculation of the weighting functions. The Optimal Estimation Method has been implemented to solve the inverse problem. The algorithm has been tested successfully on simulated spectra: it looks like one obtains a number of degrees of freedom for the aerosol retrieval around 4. The algorithm will soon be verified on real spectra, and the results will be compared with CIMEL and Brewer data.

WP 1300 Climate gases

Formaldehyde (H_2CO) is one of the most abundant carbonyl compounds and a central component of the oxidation of volatile organic compounds (VOC). It represents the total amount of oxidized VOC. Its observation can help to constrain VOCs emissions in models and supports air quality control regulations. Both NOx and VOC concentrations determine the production of ozone in the troposphere, a climate gas. Sources of H_2CO are methane oxidation (background), biogenic VOCs oxidation (isoprene), anthropogenic hydrocarbon oxidation, and biomass burning. Sinks are oxidation by OH radical and photolysis. H_2CO is a major source of CO.

Strategies have been developed to retrieve H_2CO from MAXDOAS spectra and from FTIR spectra, above Uccle and Ile de La Réunion. The MAXDOAS and FTIR inversion results for the tropospheric columns of H_2CO agree very well in capturing the diurnal, daily and seasonal variabilities. There is also good agreement with correlative GOME or SCIAMACHY data, retrieved independently using a DOAS technique. A small bias observed between MAXDOAS or SCIAMACHY satellite data and FTIR columns probably has its origin in spectroscopic uncertainties. At Jungfraujoch, where the H_2CO columns are about a factor 4 smaller than at Reunion Island, a special detection setup has lead to reliable results. Timeseries between Dec. 2005 and February 2007 show a distinct seasonal variation with a minimum in local winter. Work is ongoing to confront the observational data with IMAGES model simulations.

Methane (CH_4) is the third most important greenhouse gas, after H_2O and CO_2. Information about its isotopologues may provide indications for the origin and age of the sampled airmass.

Several successful retrieval strategies for the detection of $^{13}CH_4$ have been developed at Jungfraujoch, and have been compared among them. The recommended strategy uses 1 microwindow in the MCT detector range, and provides a good sensitivity up to 14 km altitude.

Retrievals of CH_3D are more difficult, because there are strong interferences with water vapour. Two successful retrieval strategies have been developed at Jungfraujoch. One of them, the multiwindow strategy, has already been verified successfully at Reunion Island for spectra taken at large solar zenith angles, despite very significant interferences with water vapour absorptions. The thus derived isotopic ratio δD appears to behave as expected. The second one, the multi-spectra strategy, will be investigated next.

It has also been possible to retrieve timeseries of hydrogen cyanide (HCN) column abundances at Jungfraujoch, using a strategy fitting simultaneously five HCN lines In the 3260-3310 cm^{-1} spectral region. The main HCN source is believed to be biomass burning, making this species a useful tracer of fires. Oxidation by the OH radical is among the identified sinks, while uptake by oceans has been hypothesized as the dominant removal process. The Jungfraujoch timeseries shows a strong feature due to the advection of biomass burning products in 1998. The comparison of the retrieved time series of partial columns between 7 and 20 km altitude above Jungfraujoch with correlative ACE-FTS partial columns show excellent agreement. In particular the seasonal variation, with a maximum in July, is observed identically in both timeseries. More work is needed to retrieve HCN at Ile de La Réunion, due to stronger interferences with water vapour.

Acetylene (C_2H_2) is among the nonmethane hydrocarbons (NMHCs) accessible to the FTIR remote sensing technique. As a product of combustion and biomass burning, it is emitted at the Earth surface and further transported and mixed into the troposphere. Destruction by OH is the main removal process. We have demonstrated that acetylene can be retrieved from FTIR spectra at Jungfraujoch using four lines close to 3251, 3255, 3278 and 3305 cm^{-1}. The timeseries of tropospheric columns above Jungfraujoch allows to characterize the strong seasonal variation of C_2H_2, with maximum columns generally observed around mid-February. On average, the peak-to-peak amplitude of the seasonal variation amounts to nearly 90% of the mean yearly column. The enhancement of HCN due to biomass burning that was observed in 1998, is well correlated with that observed in the C_2H_2 timeseries, as well as with the corresponding data for CO and C_2H_6. Comparisons between timeseries of partial columns between 7 and 17 km altitude from the ground-based FTIR and the satellite ACE-FTS instruments show a reasonable agreement, although the latter seems to be slightly biased high, especially during summertime. At present, the comparison covers the period early 2004 to the end of 2007. An extension in time of the ACE-FTS data set is needed to confirm these first conclusions.

WP 1400 Feasibility studies

Ethylene (C_2H_4) originates from a variety of anthropogenic (e.g. cars in urban areas) and natural (e.g. plants, volcanoes, forest fires) sources., It is difficult to detect C_2H_4 from FTIR spectra, due to its very short chemical lifetime, and in particular at the high-altitude remote site of the Jungfraujoch. However, we have demonstrated that it is possible to clearly detect larger absorptions of ethylene in Jungfraujoch FTIR spectra recorded during special events, e.g. under enhanced biomass burning, when the column is about 4 times larger than the average one.

Other feasibility studies have been initiated, looking at absorptions of HCFC-142b (CH_3CClF_2) in Jungfraujoch low sun spectra. Several retrieval strategies are under test.

WP 2000 - ACQUISITION AND EXPLOITATION OF SUPPORTING DATA

Laboratory spectroscopic measurements in support of the analysis of atmospheric spectra have been continued. The focus has been on water vapour and its isotopologues, the 9 micron region of the formic acid (H_2CO_2) absorptions and the 1–0 band of $^{13}C^{16}O$ near 2096 cm^{-1}.

Linelists of HDO (11500-23000 cm^{-1}) and D_2O (8800-9520 cm^{-1}) have been generated. Also, the experimental linelists of HDO and D_2O that cover the wide 5600-11600 cm^{-1} region have been built. They contain line positions, intensities, self- and air-broadening coefficients, and air-induced shifts and their statistical uncertainties. In the near-IR spectral region (4200-6600 cm^{-1}), a complete linelist has been produced for the $H_2{}^{16}O$, $H_2{}^{17}O$, $H_2{}^{18}O$ and HDO isotopologues. It is accessible to the scientific community through the web (http://www.ulb.ac.be/cpm). The water vapour continuum underlying the discrete lines is under investigation.

A new database for the 9 micron region of the formic acid spectrum was generated: it is now provided as an update to the HITRAN database and will be included in the forthcoming version of GEISA.

Our results for the line intensities of $^{13}C^{16}O$ are on average 1.5 % higher than those available for these lines in the HITRAN database, characterized by a 2–5 % accuracy. This is an excellent agreement, indicating that the accuracy of 1–0 band $^{13}C^{16}O$ line intensities in HITRAN is probably close to 2 %.

As seen above, our investigations have made use of ancillary and complementary data wherever useful.

WP 3000 Valorisation of the results.

The results have been presented at many occasions at international Symposia, and at the yearly NDACC UV-Vis and Infrared Working Group Meetings, and a number of refereed publications have already appeared. We expect much benefit also from the COST Action WAVACS. The laboratory results have been or will be integrated in the international spectroscopic databases HITRAN and GEISA. Our expertise is also recognized through our participation to European projects like SCOUT-O3, GEOmon and HYMN.