AT/01

AGACC I - Results

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

DURATION OF THE PROJECT
15/12/2005 – 31/12/2010

BUDGET
588.317€

KEYWORDS
Atmospheric composition change; trends; monitoring; remote-sensing; ground-based spectrometry; radio soundings; greenhouse gases; tropospheric chemistry; climate change

CONTEXT

We live in an era in which human activities are causing significant changes to the atmospheric environment which result in local to global consequences on the ecosystems. Changes in the atmospheric composition impact our climate via chemical and dynamical feedback mechanisms; in many instances they also affect air quality, and the health of the biosphere. Monitoring and understanding those changes and their consequences is fundamental to establish adequate actions for adaptation to and mitigation of the environmental changes. Furthermore, after implementation of regulatory measures like the Montreal Protocol, it is necessary to verify whether the measures are effective. This can only be achieved if we have adequate detection methods and a reliable long record of a series of key geophysical parameters.

Thus the AGACC project contributes to the provision of basic new knowledge regarding the atmospheric composition and its changes, based on advanced ground-based monitoring, in combination with satellite and numerical modelling data. Its results are integrated in ongoing international research programmes.

OBJECTIVES

The general objective of AGACC has been to improve and extend the ground-based detection capabilities for a number of climate-related target species and, based hereupon, analyse past and present observations to derive new information about the atmospheric composition, its variability and long-term changes. Despite the advent of a growing and more performant fleet of Earth Observation satellites, ground-based observations are still indispensable to (1) guarantee long-term continuity, homogeneity and high quality of the data, and (2) to underpin the satellite data for calibration and (long-term) validation.

CONCLUSIONS

A first target gas is atmospheric water vapour. It is the key trace gas controlling weather and climate. It is also the most important greenhouse gas in the Earth's atmosphere. Its amount and vertical distribution are changing, but how and why? Especially in the upper troposphere - lower stratosphere, the radiative effects of changes in the water vapour are significant and should be quantified. The measurement of water vapour is a hot topic since several years. It is a challenge, because water vapour exhibits a large gradient in its concentration when going from the ground to the stratosphere, and because it is highly variable in time and space. For example, we have found that the time scale of the variations of the total water vapour amount at Jungfraujoch is in the order of minutes.

In AGACC, we have therefore investigated various experimental techniques to measure the concentration of water vapour in the atmosphere, focusing on the total column as well as on the vertical distribution in the troposphere up to the lower stratosphere. The retrieval of water vapour vertical profiles and total columns from ground-based FTIR data has been initiated at three very different stations where correlative data for verification are available, namely Ukkel (± sea level, mid-latitude), Ile de La Réunion (± sea level, tropical) and Jungfraujoch (high altitude, mid-latitude), with promising results. In particular, at Jungfraujoch, it has been demonstrated that the precision of the FTIR integrated water vapour (IWV) measurements is of order 2%. The capability to retrieve individual isotopologues of water vapour, and to monitor their daily and diurnal variations, has also been demonstrated. This could open new ways to study in the future the role of water vapour in the radiative balance, the global circulation, precipitation etc. We also started joint exploitation of ground-based FTIR and satellite IASI data for water vapour and its isotopologues, in order to exploit fully the potential of the existing instrumentation.

AGACC I - Results

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

A correction method for the radiosoundings at Ukkel has been successfully implemented, resulting in a homogeneous and reliable time series from 1990 to 2008 from which trends in upper troposphere humidity (UTH) and tropopause characteristics have been derived. One observes a rising UTH until September 2001, followed by a decline, accompanied by a descent and heating of the tropopause up to the turning point and an ascent and cooling afterwards. The changes after September 2001 in the upper troposphere can be explained by surface heating and convective uplift. At Jungfraujoch, one does not observe any significant trend in the total water vapour abundance above the station over the 1988-2010 time period, although significant positive summer and negative winter trends have been detected.
We have made a quantitative statistical comparison between ground-based FTIR, CIMEL, GPS and integrated (corrected) radio sounding measurements of the IWV at Ukkel. This work is important to better characterize the different sensors in order to exploit together different observations made by different instruments.

A second target species is atmospheric aerosol. There is a very large variety of aerosol both from natural or anthropogenic origin. One of the reasons why they are so important is that they affect the optical properties of the atmosphere. In particular, it has been demonstrated in previous studies that the aerosols have a large impact on the quantity of harmful UV-B radiation received at the Earth's surface. The latest IPCC Report also stressed that the radiative forcing caused by atmospheric aerosols is one of the largest uncertainties in determining the total radiative forcing in the atmosphere. Better monitoring capabilities of aerosol properties can therefore improve our understanding and forecasting of the atmospheric processes and evolution, and in particular of UV-B and climate changes.

Several measurement techniques are now operational in the AGACC consortium for the ground-based monitoring of aerosol properties. These are the Brewer spectrometer and CIMEL observations at Ukkel, the latter contributing also to the AERONET network since July 2006, and the newly developed MAXDOAS observations. Unlike CIMEL and Brewer measurements, that provide the total Aerosol Optical Depth, it has been demonstrated that the MAXDOAS measurements also provide additional information about the vertical distribution of the aerosol extinction in the lowest kilometres of the troposphere.

A better understanding of the ultimate capabilities of MAXDOAS aerosol remote sensing has been gained through participation to the international CINDI campaign (Cabauw Intercomparison Campaign of Nitrogen Dioxide measuring Instruments) in summer 2009. The combination of Brewer, CIMEL and MAXDOAS instruments gives us a remote-sensing dataset that will enable a more comprehensive characterization of the tropospheric aerosol optical properties. The usefulness of these aerosol observations has already been demonstrated in the improvement of the UV-index predictions for the general public. Another application is their use as input data in the retrieval of vertical profiles of tropospheric pollutants from MAXDOAS measurements, like tropospheric NO_2 and formaldehyde.

Third we have focused on a few climate-related trace gases. Changing greenhouse gas and aerosol concentrations directly affect the radiative budget of the atmosphere, and therefore climate. But many species known as pollutants like carbon monoxide (CO), nitrogen oxides (NOx) and hydrocarbons, - often related to fossil fuel or biomass burning -, also affect climate through their role in chemical reactions that produce tropospheric ozone, which is a well-known greenhouse gas, or that modify the lifetime of gases like methane, or the oxidation capacity of the atmosphere.

Therefore in AGACC, we have focused on the measurement of a number of trace gases that are subject to changing concentrations, that directly or indirectly affect climate, and that are either difficult to monitor or that have not yet been measured from the ground. We have included attempts to observe distinctly some isotopologues, because the isotopic ratios observed in an airmass provide information on its history, and because the FTIR solar absorption measurements provide a rather unique capability hereto.

The investigated species are the isotopologues of CH_4 and CO, and hydrogen cyanide (HCN), as examples of biomass burning tracers, some hydrocarbons like formaldehyde (HCHO), ethylene (C_2H_4) and acetylene (C_2H_2), and HCFC-142b, a replacement product for CFCs and a greenhouse gas.

In many cases, retrieval strategies had to be adapted when going from one site to another with different atmospheric conditions, especially when the local humidity and abundances are very different as is the case between Jungfraujoch (dry, high altitude, mid-latitude) and Ile de La Réunion (humid, low altitude, low latitude). Still we have been able to show the feasibility of retrieving particular trace gas information even under difficult conditions. Many of our results have been compared to correlative data, to validate the approach and to gain complementary information. It is also important to note that the retrieval strategies developed in AGACC have regularly been presented to the global Network for the Detection of Atmospheric Composition Change (NDACC) UV-Vis and Infrared communities and have often been adopted by others or even proposed for adoption as a standard in the community (e.g., for hydrogen cyanide (HCN)).

In particular:

We have been able to study the seasonal variations of *HCN* at the Jungfraujoch and at Ile de La Réunion, and to show the dominant impact of biomass burning.

Formaldehyde was studied in much detail at Ukkel, Jungfraujoch and Ile de la Réunion. The challenge for detection at Jungfraujoch is the small abundance (about 10 times smaller than at Ukkel and Ile de La Réunion); a particular observation strategy was developed successfully, resulting in a time series that already shows the day-to-day and seasonal variations. At Ile de La Réunion, comparisons of FTIR, MAXDOAS, satellite and model data have (1) shown the good agreement between the various data sets, but also, (2), the variability of HCHO (diurnal, seasonal, day-to-day), and (3), thanks to the complementarities of the various data sets, they have enabled us to learn more about the long-range transport of Non-methane Volatile Organic Compounds (NMVOCS, precursors of HCHO) and deficiencies in the models. It was shown that fast, direct transport of NMVOCS from Madagascar has a significant impact on the HCHO abundance and its variability at Ile de La Réunion, and that this is underestimated in the model.

Significant progress was made as to the detection of $^{13}CH_4$ *and* CH_3D from ground-based FTIR observations, both at Jungfraujoch and Ile de La Réunion. To our knowledge, it is the first time that a $\delta^{13}C$ data set is derived from ground-based FTIR observations. More work is needed to improve the CH_3D retrieval at Ile de La Réunion, and to interpret the results, in combination with models.

Also for the first time, ^{12}CO *and* ^{13}CO have been retrieved individually at Jungfraujoch. The $\delta^{13}C$ time series shows significant seasonal and interannual changes.
As to the hydrocarbon *ethylene*, it is shown that it can be detected at Jungfraujoch only in spectra at low solar elevation, given its small atmospheric abundance.
Regarding acetylene, the observed time series at Jungfraujoch and Ile de La Réunion show clear seasonal variations and enhancements due to the impact of biomass burning events, correlated with enhancements in CO, C_2H_6 and HCN.

It is not clear yet whether we can reliably retrieve the concentration of HCFC-142b, a replacement product that is increasing strongly in the troposphere. New line parameters for the interfering species HFC-134a are required to confirm/infirm the preliminary results. This highlights again the importance of the laboratory work for providing such parameters.

Improved line parameters have been obtained for water vapour and its isotopologues, ethylene and formic acid. These AGACC results have been integrated in the international spectroscopic databases. We also showed that line intensities available around 2096 cm^{-1} for the $^{13}C^{16}O$ isotopologue of carbon monoxide in the HITRAN database seem to be accurate to 2%. We failed to improve line intensities for the 13.6 □m region of acetylene.

The new data sets that have been derived in AGACC from FTIR and MAXDOAS observations have been archived in the NDACC data centre, where they are available for users (generally modelers and satellite teams). In addition, they are stored locally and are available to users upon request.

AGACC I - Results

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

AGACC results have been reported to the international scientific community, via the literature, via integration in geophysical or spectroscopic databases, and via participation to international research initiatives like the Atmospheric Water Vapour in the Climate System (WAVACS) Cost Action, the International Space Science Institute (ISSI) Working Group on Atmospheric Water Vapour, the International Union of Pure and Applied Chemistry (IUPAC) project, the International CINDI campaign, etc.

The results have already found important scientific applications. A few examples are worth mentioning: the re-evaluation of methane emissions in the tropics from SCIAMACHY based on the new H_2O spectroscopy, and the improved retrievals of HCOOH from the satellite experiments ACE-FTS and IASI, and from the ground.

CONTRIBUTION OF THE PROJECT TO A SUSTAINABLE DEVELOPMENT POLICY

In the longer-term, the AGACC results will no doubt benefit the research in atmospheric sciences –in particular in the monitoring of its composition changes–, which is the fundamental basis of environmental assessment reports for supporting policy makers.

CONTACT INFORMATION

Website of the project:
http://www.oma.be/AGACC/Home.html

Coordinator

Martine De Mazière
Belgian Institute for Space Aeronomy (BIRA-IASB)
Ringlaan 3
B -1180 Brussels
Tel: +32 (2) 373 03 63
Fax: +32 (2) 374 84 23
martine.demaziere@aeronomie.be
www.aeronomie.be

Promoters

Hugo De Backer
Royal Meteorological Institute of Belgium (KMI-IRM)
Ringlaan 3
B-1180 Brussels
Tel: +32 (2) 373 05 94
Fax: +32 (2) 375 12 59
Hugo.DeBacker@kmi-irm.be
www.meteo.be

Emmanuel Mahieu
University of Liège (ULg)
Institute of Astrophysics and Geophysics
Groupe Infra-Rouge de Physique Atmosphérique et Solaire (GIRPAS)
Allée du 6 Août 17
B-4000 Liège
Tel: +32 (4) 366 97 86
Fax: +32 (4) 366 97 47
emmanuel.mahieu@ulg.ac.be
www.girpas.astro.ulg.ac.be

Michel Carleer
Université Libre de Bruxelles (ULB)
Service de Chimie Quantique et de Photophysique (SCQP)
Av. F. D. Roosevelt, 50 CP160/09
B-1050 Brussels
Tel: +32 (2) 650 24 25
Fax: +32 (2) 650 42 32
mcarleer@ulb.ac.be
www.ulb.ac.be/cpm

SSD SCIENCE FOR A SUSTAINABLE DEVELOPMENT

BELGIAN SCIENCE POLICY
Louizalaan 231 Avenue Louise • B-1050 Brussels
Tél. +32 (0)2 238 34 11 • Fax +32 (0)2 230 59 12 • www.belspo.be/ssd
Contact : Martine Vanderstraeten

ATMOSPHERE | CLIMATE