Name of research institute or organization:

Belgian Institute for Space Aeronomy (BIRA-IASB)

Title of project:

Atmospheric physics and chemistry

Part of this programme:

NDACC, NORS, ACTRIS, AGACC-II

Project leader and team:

Dr. M. Van Roozendael: project leader UV-Vis
Dr. Martine De Mazière: project leader FTIR
Bart Dils, Bavo Langerock, Corinne Vigouroux, Caroline Fayt, Clio Gielen, François Hendrick, Christian Hermans, Gaia Pinardi: team scientists

Project description:

UV-Vis (main results, significance of results, progress in 2014)

In 2014, BIRA-IASB has continued its long-term monitoring of atmospheric trace gases using the SAOZ instrument operated at Jungfraujoch since 1990 and the more recent MAXDOAS system in place since 2010. Although the MAXDOAS system could provide measurements during most of the time in 2014, the SAOZ instrument was damaged due to lightning in July 2014, and since then was discontinued. We are investigating the possibility of replacing the SAOZ by a new zenith-sky system of similar performance based on the new technology of the mini-spectrometers made by the Avantes company. In the meantime the continuity of both stratospheric and tropospheric trace gas observations is being ensured using the MAXDOAS instrument. Concerning the data exploitation, the main focus in 2014 has been on analyzing the tropospheric content of nitrogen dioxide (NO_2) and formaldehyde (HCHO). These studies have been performed in the context of the EU project NORS and the national project AGACC-2.

In the EU FP7 NORS project, we have been cooperating with EMPA on the integration/inter-comparison of surface in-situ observations with remote-sensing DOAS measurements. Since surface in-situ observations are very precise and can be traced back to international standards, this activity serves as a demonstration of the quality of the remote-sensing products in the troposphere. However, in order to facilitate a meaningful comparison, the representativeness of the surface in-situ and ground based remote sensing observations needs to be taken into account. A novel method was developed at EMPA that uses backward Lagrangian Particle Dispersion Modelling (LPDM) to characterise the representativeness of different types of observations. The LPDM simulations were tailored towards each specific sampling volume. They helped to 1) characterise the history of each sampled air mass (remotely sensed and in-situ), and in turn their representativeness, and 2) to generate high-resolution model profiles specific for the remote sensing volumes. The latter were then merged with the surface in-situ observations and yielded the aspired reference profiles against which the remote sensing data were validated. The analysis was carried out for FTIR observations of CO, CH4, and O3 as well as for MAXDOAS NO_2 retrievals. Virtually no bias (<20 ppt) was determined for the MAXDOAS NO_2 observations at Jungfraujoch.

In the AGACC-2 (Belgian national project) and NORS projects, HCHO profiles have been successfully retrieved from ground-based Fourier Transform Infrared (FTIR) solar spectra and UV-Visible MAX-DOAS data recorded during the July 2010 – December 2012 time period. Analysis of the retrieved products has revealed different vertical sensitivity between both remote sensing techniques. Accordingly, HCHO amounts simulated by two state-of-the-art Chemical Transport Models (CTMs), GEOS-Chem and IMAGESv2, have been successively compared to FTIR total columns and MAX-DOAS 3.6 – 8 km partial columns, accounting for the respective vertical resolution of each ground-based instrument. Using the CTMs outputs as intermediate, FTIR and MAX-DOAS retrievals have shown consistent seasonal modulations of HCHO throughout the investigated period, characterized by summertime maximum and wintertime minimum. Such comparisons have also highlighted that FTIR and MAX-DOAS provide complementary products allowing to characterise the behavior of the free-tropospheric HCHO at Northern mid-latitude.

FTIR solar absorption spectrometry (main results, significance of results, progress in 2014)

In WP4 of the EU ACTRIS project, the primary focus is the optimisation and harmonisation of the various in-situ Volatile Organic Compounds (VOC) and NOx measurements. The measurements of these species are crucial for our understanding of various atmospheric and emission processes. However, it is uncertain to what extent these measurements yield information on the total column above the site and whether they could become useful for total-column comparisons such as in the context of satellite validation. The study carried out by BIRA-IASB explores the potential value of in-situ measurements in said context by looking at comparisons with total column FTIR measurements and CTM model data. Due to constraints in FTIR/in-situ overlap, we have limited ourselves to the study of C_2H_6 and focused on the High Altitude Research Station Jungfraujoch (HFSJ) in the Swiss Alps (3580 m asl), which harbours both measurement types (FTIR Data from Liège University, in-situ data from EMPA). This study was finalized in 2014. It concluded as follows:
Source region analysis clearly showed the strong dependence of source region contribution as a function of altitude and thus the need for additional (Model) data to make an informed prediction of the total column content. IMAGES model vs. FTIR comparisons yield a higher correlation than FTIR vs in-situ comparisons, which speaks for the model quality, even if a significant bias is still present. Notwithstanding its high quality, adding in-situ information to said model does yield a further improvement of the data quality, confirming that in-situ data can indeed be useful for inferring a more accurate total column value.

The in-situ correction method employed here was designed to explore if such a correction could indeed be fruitful and given its limited purpose is therefore rather crude. A more sophisticated approach, using tracer simulations in order to infer the height till which the in-situ correction should propagate, has been developed by EMPA and could be applied in a next step (see the work done in NORS as outlined above).

In 2014, the analysis of O_3 trends (total column trends and partial column trends in 4 atmospheric layers based on FTIR data) which already started in 2012 and was extended in 2013 to non-European stations, has been finalized and published (Vigouroux et al., ACPD 2014). A multivariate regression analysis method has been applied to the data, highlighting the various processes that impact the observed O_3 variability and trends. This study has been completed as a contribution to the international SI^2N initiative[1] which coordinates the input of the international scientific community on the assessment of the variability and long-term changes in the vertical distribution of ozone.

BIRA-IASB has coordinated the EU FP7 NORS project in which Jungfraujoch is one of the 4 demonstration stations used as an input to the automated NORS Copernicus Atmospheric Service validation system (http://nors-server.aeronomie.be). The Jungfraujoch FTIR and UV-Vis data have been used extensively in the MACC-II Validation reports (see http://www.copernicus-atmosphere.eu/services/ aqac/global_verification/validation_reports/). The project ended very successfully at the end of November 2014, with an international NORS/NDACC/GAW Workshop organized by BIRA-IASB in Brussels. The NORS

[1] **SPARC/IO_3C/IGACO-O3/NDACC Activity on Past changes in the Vertical Distribution of Ozone**

Validation Server is continued in the MACC-III project, and Jungfraujoch data continue to be used for validation of the MACC-III products as soon as they are submitted to the NDACC/NORS database.

Key words:

Atmospheric composition, long-term monitoring, optical remote sensing, vertical inversion methods, satellite and model validation

Internet data bases:

- The data are archived in the NDACC database (http://www.ndacc.org/), in the NADIR/NILU database (http://www.nilu.no/projects/nadir).
- Data processed for ENVISAT validation purposes are also submitted to the ENVISAT CAL/VAL database (http://nadir.nilu.no/calval).
- Revised HDF GEOMS formats for UV-Vis DOAS and FTIR data products have been implemented at the NDACC data base, as a contribution to the NORS project.
- In the framework of NORS, a Rapid-Delivery submission system has been implemented for several NDACC sites (among them Jungfraujoch), by which measurements are provided to the data base within 1 day to 1 month after data acquisition.

Collaborating partners/networks:

- Collaborations with University of Liège and NDACC partners
- Collaboration with European FTIR and UV-Vis teams and modelling teams in the frame of the EU project NORS
- Collaboration with M. Chipperfield of Univ. Leeds
- Both the UV-Vis and FTIR observations contribute to the international Network for the Detection of Atmospheric Composition Changes (NDACC)
- Collaboration with B. Buchmann, D. Brunner, S. Henne, S. Reimann and M. Steinbacher of EMPA (NORS and ACTRIS projects)
- Collaboration with F. Goutail, J.-P. Pommerau and A. Pazmino of LATMOS, France (SAOZ)
- Collaboration with the OMI, ACE and MetOp GOME-2 and IASI satellite communities
- Collaboration with Université Libre de Bruxelles for IASI FORLI data validation
- Collaboration with S&T for the NORS Validation Server

Scientific publications and public outreach 2014:

Refereed journal articles and their internet access

Franco, B., F. Hendrick, M. Van Roozendael, J.-F. Müller, T. Stavrakou, E.A. Marais, B. Bovy, W. Bader, C. Fayt, C. Hermans, B. Lejeune, G. Pinardi, C. Servais, and E. Mahieu, Retrievals of formaldehyde from ground-based FTIR and MAX-DOAS observations at the Jungfraujoch station and comparisons with GEOS-Chem and IMAGES model simulations, Atmos. Meas. Tech. Discuss., **7**, 10715-10770, doi: 10.5194/amtd-7-10715-2014, 2014.
http://www.atmos-meas-tech-discuss.net/7/10715/2014/amtd-7-10715-2014.html

Gielen, C., M. Van Roozendael, F. Hendrick, G. Pinardi, T. Vlemmix, V. De Bock, H. De Backer, C. Fayt, C. Hermans, D. Gillotay, and P. Wang, A simple and versatile cloud-screening method for MAX-DOAS retrievals, Atmos. Meas. Tech., **7**, 3509-3527, doi: 10.5194/amt-7-3509-2014, 2014.
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Hassler, B., I. Petropavlovskikh, J. Staehelin, T. August, P. K. Bhartia, C. Clerbaux, D. Degenstein, M. De Mazière, B. M. Dinelli, A. Dudhia, G. Dufour, S. M. Frith, L. Froidevaux, S. Godin-Beekmann, J. Granville, N. R. P. Harris, K. Hoppel, D. Hubert, Y. Kasai, M. J. Kurylo, E. Kyrölä, J.-C. Lambert, P. F. Levelt, C. T. McElroy, R. D. McPeters, R. Munro, H. Nakajima, A. Parrish, P. Raspollini, E. E. Remsberg, K. H. Rosenlof, A. Rozanov, T. Sano, Y. Sasano, M. Shiotani, H. G. J. Smit, G. Stiller, J. Tamminen, D. W. Tarasick, J. Urban, R. J. van der A, J. P. Veefkind, C. Vigouroux, T. von Clarmann, C. von Savigny, K. A. Walker, M. Weber, J. Wild, and J. Zawodny, SI2N overview paper: ozone profile measurements: techniques, uncertainties and availability, Atmospheric Measurement Techniques, **7**, 5, 1395-1427, doi: 10.5194/amt-7-1395-2014, 2014.
http://www.atmos-meas-tech.net/7/1395/2014/

Langerock, B., M. De Mazière, F. Hendrick, C. Vigouroux, F. Desmet, B. Dils, and S. Niemeijer, Description of algorithms for co-locating and comparing gridded model data with remote-sensing observations, Geoscientific Model Development Discussions, **7**, 6, 8151-8178, doi: 10.5194/gmdd-7-8151-2014, 2014. http://www.geosci-model-dev-discuss.net/7/8151/2014/gmdd-7-8151-2014.html

Tack, F., F. Hendrick, F. Goutail, C. Fayt, A. Merlaud, G. Pinardi, C. Hermans, J.-P. Pommereau, and M. Van Roozendael, Tropospheric nitrogen dioxide column retrieval based on ground-based zenith-sky DOAS observations, submitted to AMT (2014).

Van Geffen, J. H. G. M., K.F. Boersma, M. Van Roozendael, F. Hendrick, E. Mahieu, I. De Smedt, M. Sneep, and J.P. Veefkind, Improved spectral fitting of nitrogen dioxide from OMI in the 405–465 nm window, Atmos. Meas. Tech. Discuss., **7**, 10619-10671, doi: 10.5194/amtd-7-10619-2014, 2014.
http://www.atmos-meas-tech-discuss.net/7/10619/2014/amtd-7-10619-2014.html

Vigouroux, C., T. Blumenstock, M. Coffey, Q. Errera, O. García, N. B. Jones, J. W. Hannigan, F. Hase, B. Liley, E. Mahieu, J. Mellqvist, J. Notholt, M. Palm, G. Persson, M. Schneider, C. Servais, D. Smale, L. Thölix, and M. De Mazière, Trends of ozone total columns and vertical distribution from FTIR observations at 8 NDACC stations around the globe, Atmospheric Chemistry and Physics Discussions, **14**, 17, 24623-24666, doi: 10.5194/acpd-14-24623-2014, 2014.
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