The Earth Observing System: Synergy between Ground-Based and Satellite Observations, and Model Experiments.

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ABSTRACT

The Integrated Global Observing Strategy (IGOS, http://www.unep.ch/earthw/igos.htm) is an international initiative, calling for an integrated use of observations from a multitude of platforms (ground-based, balloon, aircraft and satellite), a variety of numerical modelling efforts, and distributed data and information services. Examples are the Earth Science Enterprise (ESE, http://eospso.gsfc.nasa.gov/eos_homepage/misc_html/fact_toc.html), the Global Climate Observing System (GCOS, http://193.135.216.2/web/gcos/gcoshome.html) and the Earth Observing Science Data and Information System (EOSDIS, http://spsosun.gsfc.nasa.gov/eosinfo/EOSDIS_Site/Index. html)

Belgium has been contributing to the Network for Detection of Stratospheric Change (NDSC, http://www.ndsc.ws), an international network of ground-based observations, since the conception of the network. More recently it has investigated the exploitation of NDSC data in synergy with satellite observations and numerical model simulations. Laboratory measurements of the spectroscopic characteristics of molecules of atmospheric interest are carried out in support of the field observations.

These activities are carried out through the Belgian 'Second Multiannual Scientific Support Plan for a Sustainable Development Policy (SPSD) - Global Change, Ecosystems and Biodiversity', DWTC/ESA Prodex programmes and EC Research and Development projects. The present report focuses on the results

obtained in ESAC, Experimental Studies of Atmospheric Changes, a project in the frame of SPSD-I, that is being continued under SPSD-II (2001-2004).

INTRODUCTION

The composition of the Earth's atmosphere is changing as the concentrations of a number of radiatively and chemically active atmospheric constituents emitted at the surface are rapidly increasing, mainly due to man-made activities. These increased emissions influence atmospheric ozone, the Earth's radiative balance, hence climate, and modify the oxidising capacity of the atmosphere. It has become clear also that the upper troposphere / lower stratosphere is a region of high interest, because it controls the exchange of gases between the troposphere, where the sources are, and the stratosphere, and it controls to a great extent the radiative balance of the atmosphere.

A better knowledge of atmospheric composition and chemical processes, from the ground up to the stratosphere, is fundamental to assess the present state and changes and predict the future evolution of the Earth's environment, so that regulatory decisions about mankind practices can be identified on the basis of firm scientific grounds. This can only be achieved by a comprehensive series of complementary measurements including ground-based observations, aeroplane and balloon-borne campaigns, and global satellite missions, by improving the probing capabilities in the troposphere, and by comparing and integrating the observational data with numerical models.

ESAC has contributed to the objectives of investigating the behaviour of ozone and many key stratospheric and tropospheric species at four stations in Europe, complemented by global satellite observations. This investigation has included long-term monitoring and the evaluation of radiative, dynamical, and chemical mechanisms underlying the observed variabilities and changes. The four stations are Ukkel (Belgium, 50.5°N, 4.3°E), Jungfraujoch in the Swiss Alps (46.5°N, 8°E), Harestua (Norway, 60.2°N, 10.7°E), and Observatoire de Haute Provence (France, 44°N, 6°E); they are part of the international Network for Detection of Stratospheric Change. Additionally, ESAC was tasked to improve the acquisition of tropospheric data, and in particular, to pursue the analysis of earlier ATMOS/Space Shuttle observations to assess the possibilities to study the upper troposphere by infrared remote sensing from space (Zander and Mahieu, *this volume*). Long-term monitoring of the spectral UV irradiance has been performed at Ukkel, in compliance with the international quality standards. The atmospheric observations have been supported by numerical modelling of the atmosphere and by laboratory experiments that provide the fundamental spectroscopic and radiative data needed in the spectral data analyses and in the models.

The research is embedded in various European and international research programmes.

Partnership

Four Belgian research groups, who work in close collaboration with international partners, carried out the ESAC project. They are the Royal Meteorological Institute of Belgium, the "Groupe Infra-Rouge de Physique Atmosphérique et Solaire" of the Université de Liège, the "Laboratoire de Chimie Physique et Moléculaire" of the Université Libre de Bruxelles, and the Belgian Institute for Space Aeronomy that coordinated the project. A close collaboration among the partners was achieved through the sharing of manpower and instruments for performing observations, and the exchange of data and expertise concerning the analysis and interpretation of measurements.

Results

The ESAC project has enabled the Belgian partners to continue long-term monitoring of the atmospheric composition with ground-based instruments at the four European stations mentioned above. The instruments used are Fourier transform infrared (FTIR) spectrometers, UV-visible Differential Optical Absorption Spectroscopy (DOAS) instruments, UV-visible radiometers including Dobson and Brewer instruments and ozone sondes. Existing time series of atmospheric data have been extended and updated, in a consistent way. Revisions and homogenisations have been implemented where necessary. The synergy with satellite data has been explored. Accurate laboratory data of relevance to the atmospheric research have been acquired.

Jungfraujoch long-term observations

At the Jungfraujoch, time series of nearly two dozen atmospheric constituents date back to 1985, with some acquired since the mid seventies, even since 1950. They have proven to be of utmost importance to identify and quantify variabilities and long-term changes of a large number of atmospheric species, among which ozone, key stratospheric species that are involved in the processes leading to ozone destruction, like the halogens, a number of radiatively active gases (the so-called greenhouse gases[1]) that impact the Earth's climate, like methane and carbon dioxide, and source gases emitted at the surface, often by human activities, like the CFC and HCFC. Important features in the atmosphere's evolution that could be detected at the Jungfraujoch and investigated during the ESAC project are:

- The turn around of the rate of change of the total inorganic chlorine loading in the stratosphere, in 1997-1998. Knowing that it takes 3 to 5 years for the long-lived chlorine-bearing source gases to leave the troposphere and reach the mid-stratosphere, this observation is in agreement with the observed decrease in the total organic chlorine in the troposphere (that is representative of the chlorine-bearing source gases) after mid-1994, as shown in Figure 1.

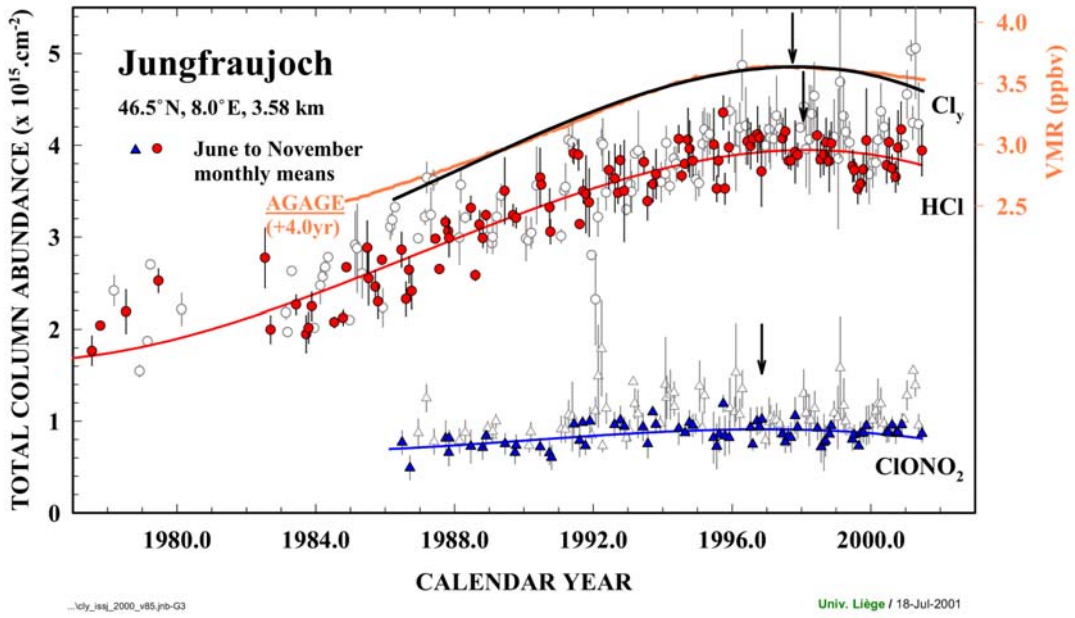

Figure 1. The timely evolution of inorganic chlorine (Cl_y) above the Jungfraujoch based on monthly mean vertical column abundances of HCl and $ClONO_2$. The maximum Cl_y loading is found here to have occurred near the end of 1997, about 4 years after the load of organic chlorine peaked in the troposphere (curve labelled AGAGE).

The observations agree with model calculations that take into account the reductions of emissions imposed by the Montreal Protocol and its successive Amendments and Adjustments. The confrontation of the evolution of the total inorganic chlorine content observed at the ground with the stratospheric burden observed by the satellite experiment HALOE and the load in organic chlorine observed by *in situ* experiments will provide important information regarding transport and troposphere-stratosphere exchanges.

[1] We use the term greenhouse gases because of its widespread use. However it must be noticed that the term is badly chosen, because the atmospheric warming effect is a physical process that is different from the one that controls the real greenhouse: the latter is based on the reduction of convective transport, while the atmospheric warming is caused by absorption of thermal infrared radiation (Kockarts, G : Aeronomie: Physique et Chimie de l'atmosphere. De Boeck. (Ed), Bruxelles, 2000).

- The continued rise, although slowed down, of the inorganic fluorine concentration in the stratosphere, contrary to the decrease of chlorine. At present this observation cannot be understood, in comparison with model predictions and correlative observations.
- The decrease of the NO_2 column abundance by about 45% at the beginning of 1992, due to large amounts of aerosols injected in the stratosphere by the eruption of the Mt. Pinatubo volcano in the Philippines in July 1991, and its subsequent recovery by the end of 1994.
- The now apparent long-term increase of NO_2 by about 0.6%±0.2% per year. At present, the trends of HNO_3 (-0.2%±0.2% per year) and NO_2 don't seem to be consistent with the one observed for the N_2O source gas, that is of order 0.30%±0.01% per year. This is related to the difficulty to quantify the long-term changes, due to the very large natural variability of HNO_3 and the perturbation by Mt. Pinatubo aerosols that disrupted the time series.
- Observations of the rates of increase of the major radiatively active gases that are to be controlled by the Kyoto Protocol: CO_2 (0.41%±0.01% / year), CH_4 (slowing down from 0.74%/year in 1987 to 0.1%/year in 2000), N_2O (0.30%/year), SF_6 (slowing down from 14%/year in 1987 to 5%/year in 2000).
- Tropopause heights appear to have risen during the eighties, and lowered in the nineties. This may correlate with changes in the radiative balance in the atmosphere. The tropopause changes may also explain observed differences in the rates of change of N_2O sea-level concentrations and total columns; they may also correlate with the slowing in the nineties of the negative total ozone trend that was observed in the eighties. Has a recovery of ozone been observed after 1994 ? The question whether this signature is due to interannual variability or to a steady trend will be answered by extending the times series in the future.

The evolution of Ozone at Ukkel

The time series of ozone vertical profile data between 0 and 35 km altitude, obtained from soundings at Ukkel, extends from 1969 to present. It has been re-evaluated to correct for instrumental artefacts and changes with time. Trends have been revised concurrently. The main features are: a pronounced long-term decrease of ozone in winter and spring in the lower stratosphere, an increase of tropospheric ozone in all seasons, and a strong increase of ozone in the planetary boundary layer in March to September and decrease during the other months. All-year round trend values for ozone are: positive in the troposphere of order +0.35 to 0.85%/year (depending on altitude), negative in the lower stratosphere between –0.2 and –0.5%/year (at 15 km altitude). It appears that the photochemical production of ozone has increased significantly during recent years.
A comparative study of the re-evaluated ozone sounding data with co-located ozone profiles obtained from space by the SAGE-II instrument has confirmed the homogeneity of the Ukkel data. An example of the results is shown in Figure 2.

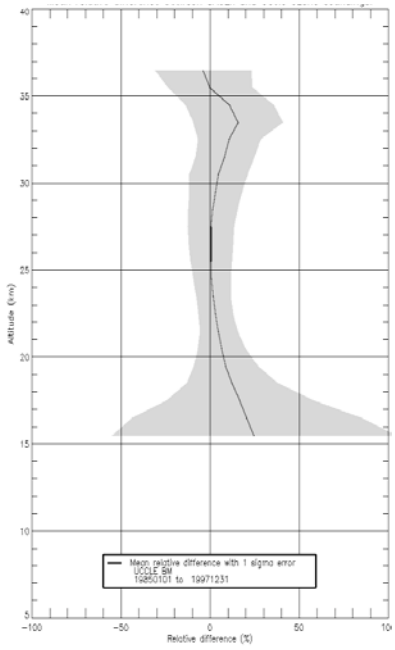

Figure 2. Mean relative difference between collocated ozone profiles from SAGE II and from Brewer-Mast ozone sonde launches at Ukkel.

OClO and BrO observations at Harestua and OHP

Growing time series of BrO at Harestua and OHP and of OClO at Harestua have improved our knowledge about the diurnal, seasonal and latitudinal variations of both species. Events of enhanced OClO values are observed in cold winters when vortex air passes over Harestua. Largest BrO amounts are observed in winter, especially under conditions of denoxification for which the conversion to the reservoir $BrONO_2$ is reduced.

The synergistic exploitation of the ground-based, balloon-borne and satellite (GOME/ERS-2) observations of BrO has led to the conclusion that there exists a non-negligible tropospheric background of BrO, on a global scale. Its sources are still to be identified. As demonstrated in Figure 3, assuming a tropospheric amount in the background spectrum leads to good agreement between ground-based and GOME BrO columns, whereas the assumption of a purely stratospheric BrO profile leads to unrealistic BrO columns for the ground-based instruments at both stations.

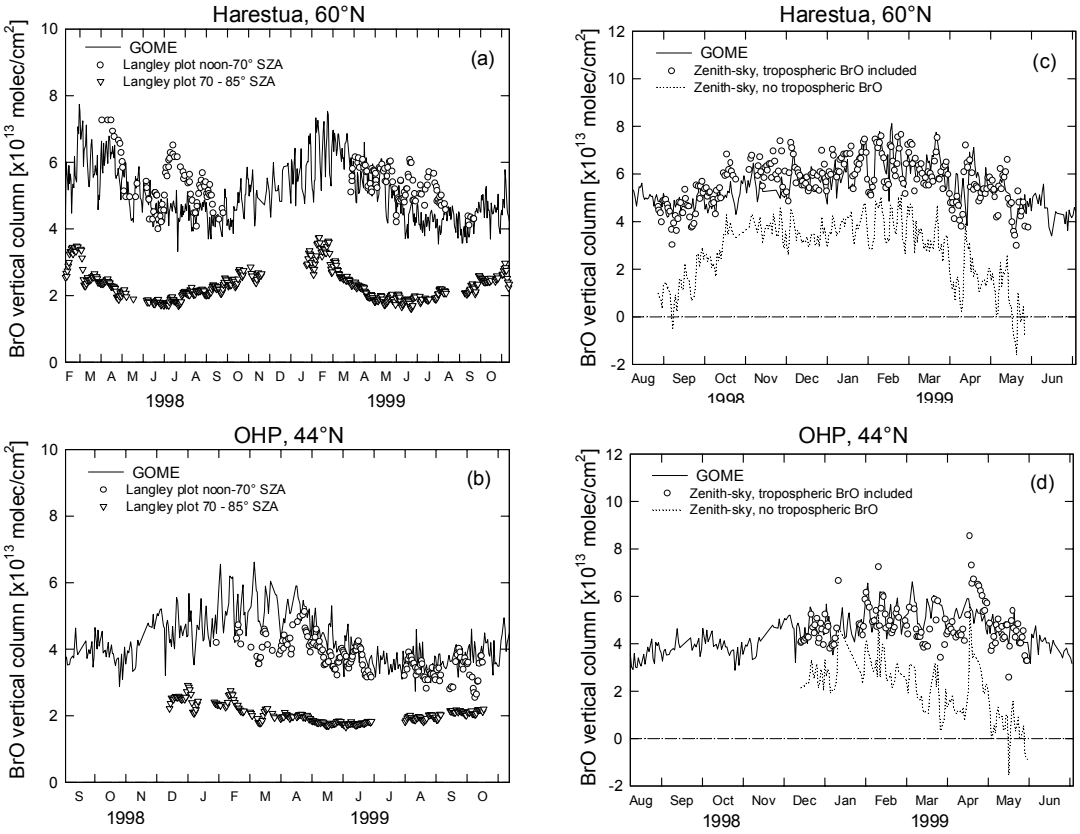

Figure 3. GOME BrO vertical columns calculated in 500 km radius around indicated ground-based stations, compared to vertical columns derived from zenith-sky data by a Langley plot method (left panel) or from noon "GOME overpass" analyses using a seasonal reference spectrum (right panel).

Using the coupled radiative transfer and photochemical box model PSCBOX (see below) for comparisons with observations, our actual knowledge of the halogen chemistry has been questioned, because twilight observations of OClO under weak chlorine activation conditions cannot be reconciled with the PSCBOX model results. More investigations are in progress.

UV spectral irradiance measurements at Ukkel

Having acquired now about 11 years of UV spectral irradiance data, UV climatology has been developed. The key factors that influence the UV dose at ground level are the occurrence of clouds and the amount of

ozone: their impact has been studied. Because of the large variability of these factors, the available time series of UV data appears to be too short to detect a possible UV trend in a reliable way.

An operational UV index forecasting procedure has been developed and implemented: UV Index forecasts are now disseminated daily to the public in the late spring and summer months.

Synergy with satellite data

The consistencies and complementarity between various satellite, balloon borne, ground-based remote sensing and *in situ* data sets have been investigated and exploited, in particular for the derivation of an NO_2 profile climatology from the ground up to 70 km altitude (Lambert et al., this volume) and for the verification of an improved algorithm for the ATMOS/Shuttle experiment extending the derived molecular profiles down into the free troposphere (Zander and Mahieu, this volume).
As an example, Figure 4 shows the comparison between NO_2 vertical column data from ground-based UV-Vis SAOZ and FTIR instruments and the HALOE experiment on board UARS, observed at the Jungfraujoch between November 1991 and December 2000. The investigation of the observed discrepancies is still in progress; part of the differences between HALOE and SAOZ data may originate in the presence of a non-negligible tropospheric NO_2 amount to which the SAOZ measurements are sensitive but not the HALOE measurements.

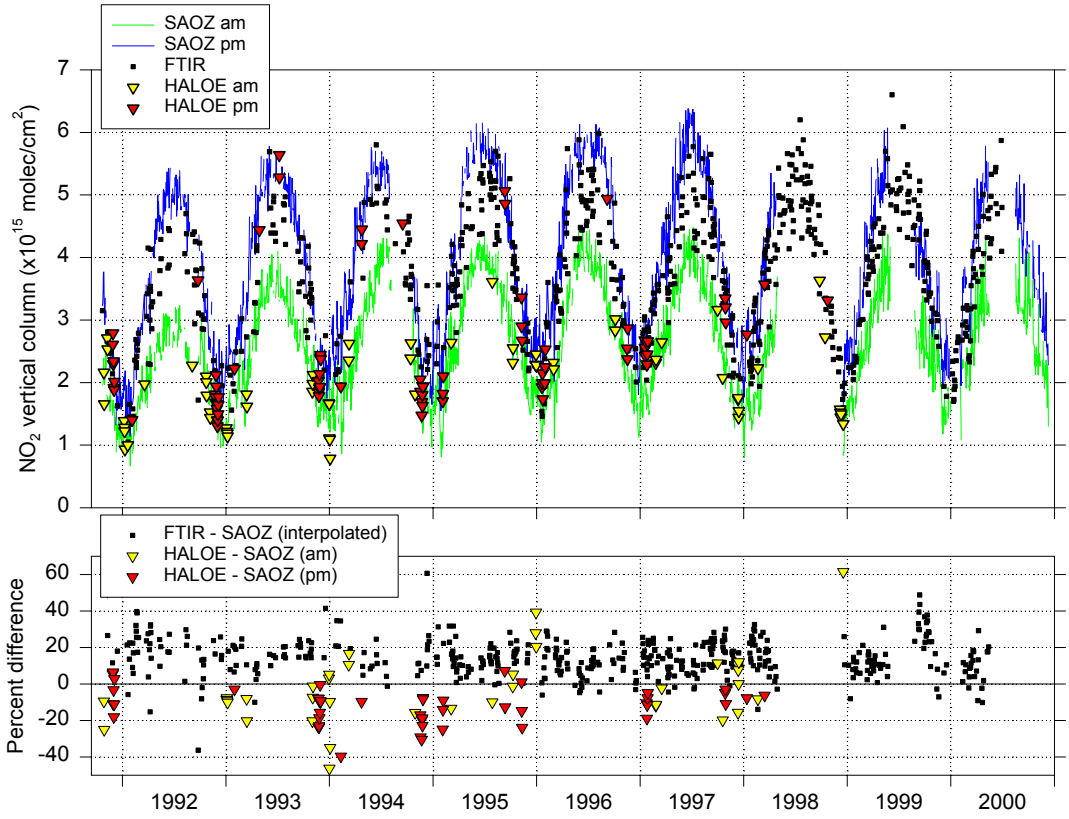

Figure 4. NO_2 total column measurements by SAOZ, FTIR and HALOE instruments above the Jungfraujoch between November 1991 and December 2000 (upper plot). The relative differences between the various data sets and SAOZ measurements are displayed in the lower plot.

Process Studies

Several campaigns have been conducted, to study tropospheric and planetary boundary layer ozone, at Ukkel, the occurrence of ozone and other pollutants (NO_2, SO_2, benzene, toluene, formaldehyde, night time NO_3) at ground level in the urban area of Brussels (1997) and the chemical composition in the troposphere – planetary boundary layer in summer 1998 in the Jungfraujoch area.

Four components affecting the ozone budget in the boundary layer (up to 2km on average) could be identified and quantified: accumulation in the layer due to local production, transfer from the boundary layer to the free troposphere, deposition at the surface and horizontal advection. It has been found that good estimates of the O_3 concentration can be made using only meteorological parameters (wind speed and direction, humidity, irradiance, …), but that they can be improved upon including estimates of NO and NO_2 concentrations.

The measurements in the Jungfraujoch area were mainly of technological interest, proving the new measurement concept of altitude-differential measurements (see below). Fast variations of CO with a 1-hour timescale have been observed, in agreement with correlative local *in situ* observations. Also tropospheric boundary layer C_2H_6 varies rapidly from day to day, whereas CH_4 and N_2O are quite stable in a 1-month time frame.

Laboratory data

The molecules for which new or more accurate spectroscopic data (absorption cross-sections, line positions and intensities) have been obtained are: O_2 and its collision complexes O_2-X with X=N_2, Ar, or O_2 itself), NO_2 and its dimer N_2O_4, H_2O and its isotopomers HOD and D_2O, C_2H_2, OCS, HOCl and HCFC-22 and HFC-152a.
In particular for the water molecule, it is important to characterize the large amount of weak absorption features in the UV-visible to identify their contribution to the radiative balance of the atmosphere.

The spectroscopic data were measured under different conditions of temperature and pressure in the near and mid infrared, visible and UV ranges using Fourier transform spectroscopy. Compared to previous studies, the aim in many cases was to acquire data at pressure, temperature and concentration conditions that are 'atmosphere-like', and/or to resolve discrepancies existing in actual literature data. As an example, figure 5 shows laboratory measurements of NO_2 cross sections, and their dependence on temperature (T), that was quantified by a linear regression expression to an accuracy of about 10%: $\sigma_{NO2} = \sigma_0 + a.T$.

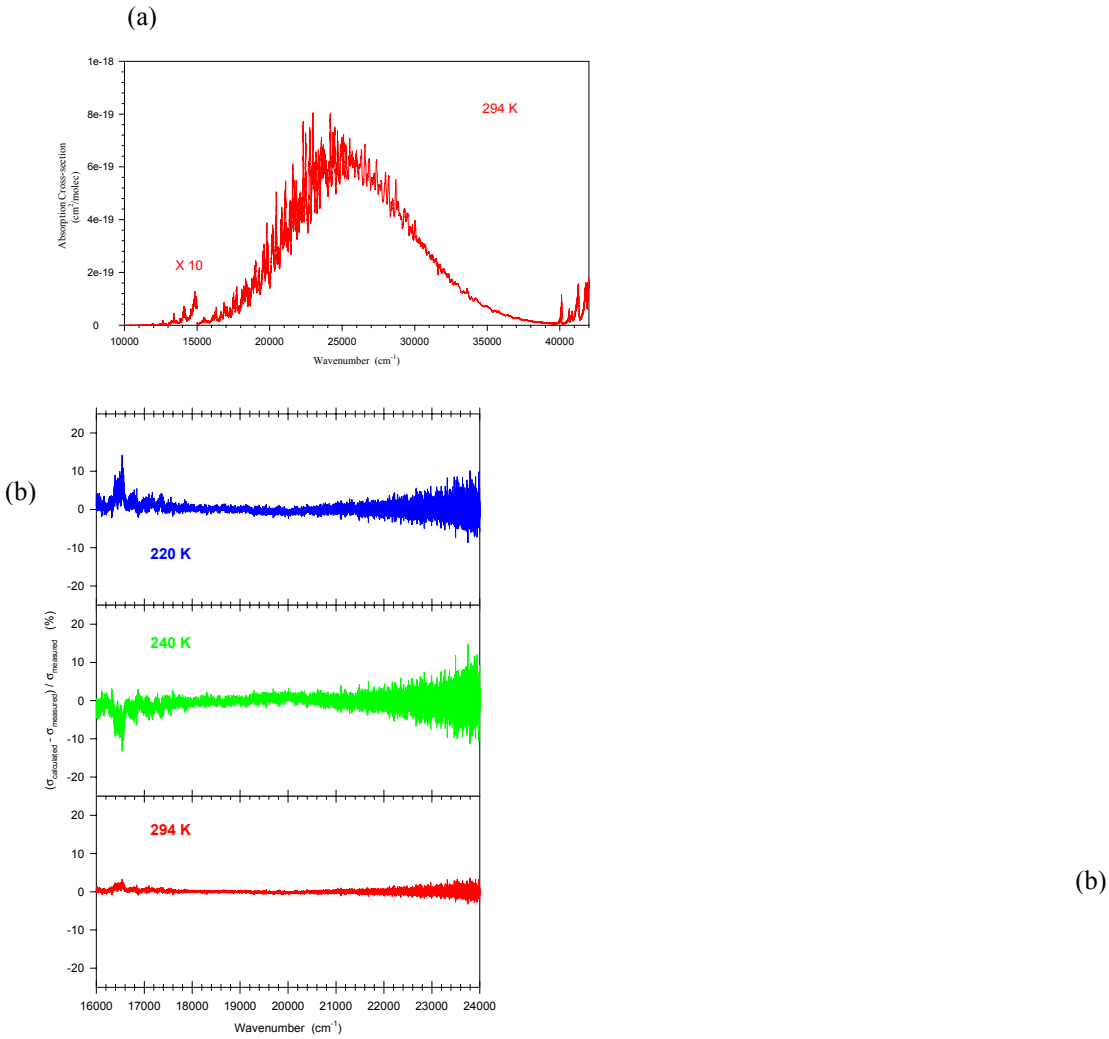

Figure 5. (a) NO_2 absorption cross-section at 294 K and at a resolution of 2 cm^{-1}. (b) Temperature effect: Comparison between observed and calculated cross sections using the linear regression parameters.

Progress in instruments, data analysis and interpretation

To achieve the objectives and to obtain the above results, various new developments and improved methods have been implemented. We cited already the *UV Index forecasting* and the *differential-altitude measurements*. The latter is one method to acquire data in a distinct altitude range in the boundary layer – lower troposphere. It is based on simultaneous measurements at two different observation altitudes of the total column abundance of a target species, to derive from the difference between the observed columns the concentration in the layer between both observation altitudes.

Vertical inversion algorithms provide an alternative method to acquire altitude information from ground-based measurements. One has been developed for high-resolution Fourier Transform infrared measurements like the Jungfraujoch observations. The principle of the inversion is the variation of the absorption line shape with altitude, due to its dependence on pressure and temperature. The inversion algorithm has been validated extensively for ozone. Preliminary results have been obtained for HCl and HF; a preliminary example for HF in comparison with correlative HALOE data is shown in Figure 6.

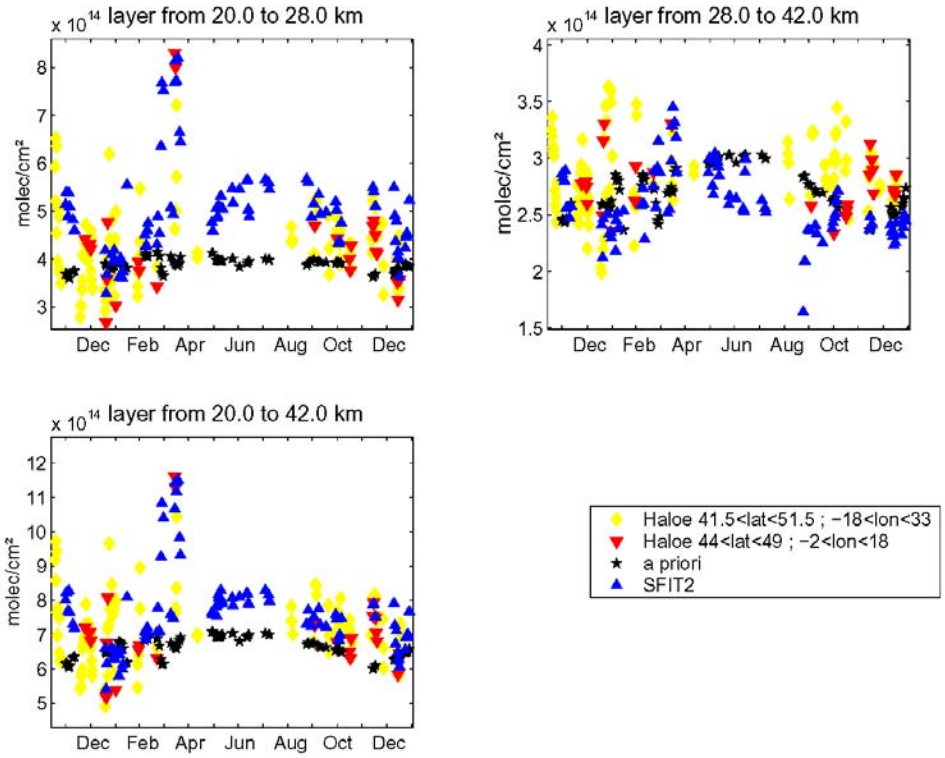

Figure 6. Comparison between HF partial column abundances from FTIR measurements at ISSJ (SFIT2, blue upward triangles) and correlative HALOE observations (yellow diamonds and red downward triangles, for more or less restricted spatial coincidence, respectively), in 2 stratospheric layers (upper left: 20-28 km, and upper right: 28-42 km) and for the total layer between 20 and 42 km (lower left).

It will be possible in the near future to re-analyse existing long-term series to study the distinct behaviour of the atmospheric composition in various altitude ranges back to the last decades. In particular, the retrieval of free tropospheric data from FTIR spectral data is very important for existing and near-future tropospheric satellite experiments like MOPITT and ENVISAT that are lacking data for validation. They will also be very beneficial for the verification of tropospheric models.

In the UV-visible DOAS method, the tropospheric information content increases if one makes quasi-simultaneous observations of the diffuse sunlight scattered at different zenith angles in the sky, e.g., the zenith and an angle close to the horizon. This is called the *DOAS off-axis method*. New instruments have been built and made operational, and preliminary results as to tropospheric BrO and formaldehyde have been obtained at the Observatoire de Haute Provence.

Software developments have contributed to a better and easier analysis of atmospheric and laboratory spectra. *WinDOAS* is a much advanced software package for the analysis of UV-visible DOAS spectra; *Wspectra and bFit* serve the analysis of absorption line parameters in high resolution Fourier transform laboratory spectra.

PSCBOX is *a coupled radiative transfer/chemical box model* that has been developed for the geophysical interpretation of fast varying species like OClO and BrO: it has contributed to a better understanding of the twilight chlorine and bromine chemistry in the stratosphere. It has also been used to simulate time series of observed NO_2 differential slant column densities (DSCDs) at various ground-based stations. Figure 7 shows a good agreement between calculated and observed NO_2 DSCDs for 90 minus 80° solar zenith angle at Harestua and OHP until August 1998. The disagreement observed from September 1998 onwards may be due to problems with the UKMO meteorological analyses that are used in the initialisation of the model.

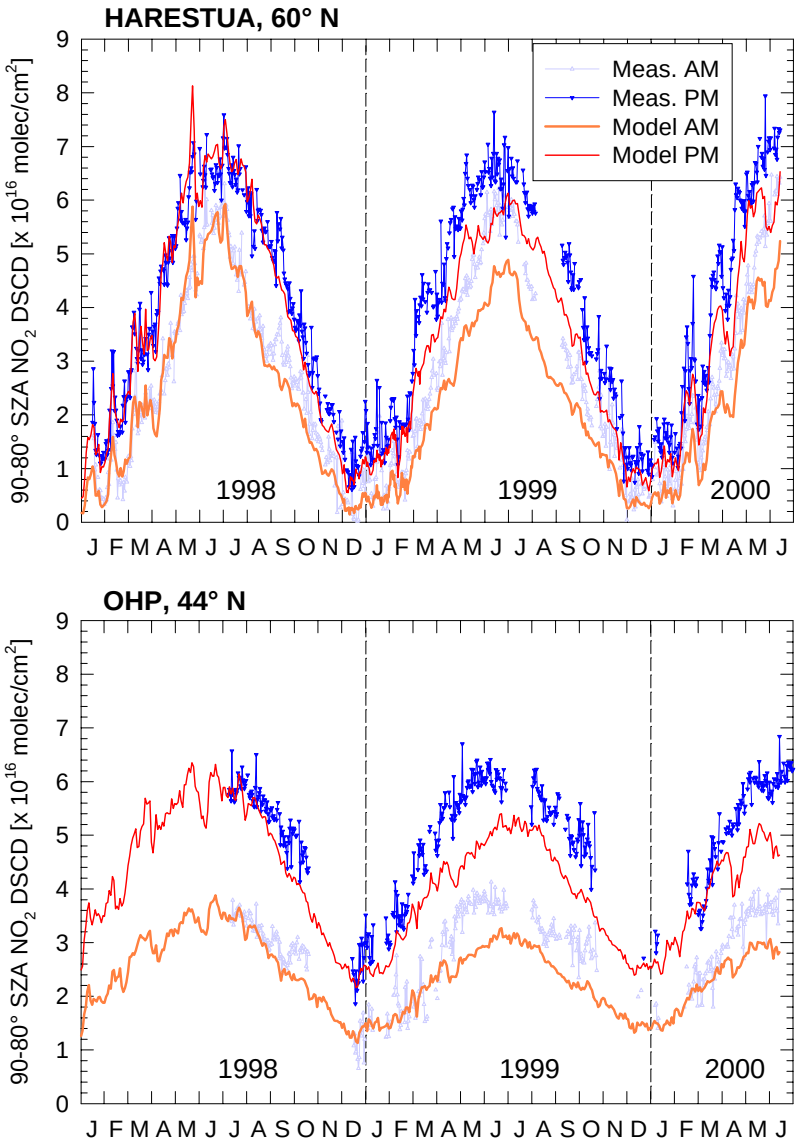

Figure 7. Time-series of measured and calculated 90-80° SZA NO_2 DSCDs at Harestua (60°N) and OHP (44°N) for the period from January 1998 through June 2000.

A new correction procedure has been developed for the ozone sonde measurements, that takes into account various correction factors. In particular, a new correction profile for the sonde pump efficiency has been implemented. The whole O_3 time series of soundings at Ukkel has been homogenised considering this new procedure.

It has been found also that reference standard atmosphere models commonly used in the community are outdated, incomplete, or lacking information about the natural seasonal and latitudinal variations. In several occasions, new climatological models have been developed for particular purposes, e.g., for O_3, based on the Ukkel soundings and local tropopause altitudes, for NO_2, based on the synergy between satellite and ground-based data and models, and for HF, HCl and CH_4, based on HALOE satellite data.

Valorisation

The usual dissemination channels have been used extensively: publications, international symposia, workshops, public information activities, integration in various European and international projects. Particular valorisation activities to be mentioned are the archiving of atmospheric and spectroscopic data in international geophysical and spectroscopic databases, respectively, via which these data become accessible to the worldwide scientific community for further exploitation including satellite validation activities, the participation of some partners in International Assessment exercises, and the distribution of software (WinDOAS) to other research groups. It has been demonstrated once more that the uninterrupted continuation of long-term monitoring supported by fundamental research (e.g., laboratory work) plays a key role in atmospheric research focusing on global changes in atmospheric chemistry, dynamics and climate.

CONCLUSIONS

The activities decribed herewith focus on ground-based remote sensing of the Earth atmosphere. The full potential of these observations is enhanced significantly when exploited in synergy with observations from satellites, *in situ* measurements, model simulations and laboratory support, as shown in this paper. Therefore, the described activities are inline with the IGOS concepts, that are the baseline for any observing system that aims at unravelling the complexity of the earth atmosphere chemistry and physics.

FUNDING SOURCES

- OSTC support through SPSD-I project ESAC, contracts CG/DD/01A to CG/DD/01D
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