2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

SESSION 7

INTERACTIONS BETWEEN ATMOSPHERIC COMPOSITION AND CLIMATE

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Oral Session

7-1 Advanced Global Atmospheric Gases Experiment (AGAGE): An NDACC Cooperating Network

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We review recent accomplishments within the multinational, global atmospheric measurement program entitled Advanced Global Atmospheric Gases Experiment (AGAGE). Using gas chromatographs (GC) with mass spectrometric (MS), electron capture (EC), flame ionization (FI) or HgO-reduction (MR) detectors (D), AGAGE measures all the important source gases for ozone-depleting atoms and free radicals covered by the Montreal Protocol and all of the important non-CO2 greenhouse gases included in the Kyoto Protocol (see Figure for sample measurements). This in situ, real-time, high-frequency surface measurement network combines boundary-layer measurements and 3D modeling to determine optimally the global rates of emission and/or destruction (i.e. lifetimes) of anthropogenic chlorocarbons, chlorofluorocarbons (CFCs), bromocarbons, hydrochlorofluorocarbons (HCFCs), hydrofluorocarbons (HFCs) and perfluorinated compounds (PFCs, and SF6) that contribute most of the reactive halogen to the stratosphere and/or are strong infrared absorbers. It also documents accurately the global distributions and temporal behaviors of biogenic/anthropogenic gases important in climate change and/or ozone depletion including methane, nitrous oxide, carbon monoxide, hydrogen, methyl chloride and methyl bromide. Recent accomplishments include measurement of new powerful greenhouse and ozone-depleting gases (e.g. SO2F2, NF3, C4F10, C5F12, C6F14, C7F16 and C8F18) and reconstruction of their past histories from in situ measurements and analyses of AGAGE and other air archives. We also determine optimally the average concentrations and trends of the tropospheric hydroxyl radical (OH) from the measured rates of destruction of atmospheric methyl chloroform, HFCs and HCFCs, combined with estimates of their anthropogenic emissions. We combine these OH estimates with global and regional measurements of methane, NMHCs, carbon monoxide and hydrogen, to test theories of primary atmospheric oxidation pathways at mid-latitudes and in the tropics. We also determine optimally, from atmospheric observations and estimates of their destruction rates, the magnitudes and distributions by region of surface sources and sinks of all of the measured gases. Finally, AGAGE provides accurate data on the global accumulation of all of these trace gases, which are used to test the synoptic, regional and global-scale circulations predicted by three-dimensional models.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Oral Session

7-2 Analysis of historical grating spectra: Jungfraujoch atmospheric database extended back to 1977

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Historical solar spectra recorded at the Jungfraujoch station (Swiss Alps, 46.55°N, 7.98°E, 3580 m a.s.l.) with a high-resolution grating spectrometer have been re-analyzed to derive total columns of a series of atmospheric gases. This instrument, built and operated by the University of Liège (Belgium), was used in the Sixties and Seventies to record two solar spectrum atlases extending from the near-ultraviolet to the near-infrared (from ~300 to ~1200 nm) [1][2]. From 1977 to 1989, it was also regularly used to record narrow spectral intervals in the mid-infrared, encompassing absorption lines of gases of atmospheric interest, e.g. CH4, HF, HCl, H2O, N2O, NO2, C2H6, O3 and CO [6]. More than 10 thousand spectra were recorded during this period.

Despite their small spectral extent (typically 5 to 10 cm-1), these grating observations may contain interesting lines belonging to gases different from the target gas, particularly numerous CH4 and H2O lines, which can then be used to increase the number of retrieved columns. Special care must however be taken to ensure that the different lines used to retrieve the columns of a given gas provide consistent results. Fortunately, from 1984 to 1989, the grating spectrometer was often operated simultaneously with a co-located home-made Fourier Transform infrared (FTIR) spectrometer, providing a convenient way to intercompare columns retrieved from different spectral ranges or derived from the same line but with 2 different instruments.

In the 1970s – 1980s, the plotted grating spectra were analyzed by manually measuring the equivalent width of the target gas lines with a high-precision Coradi planimeter [5]. They were never analyzed with fitting programs, which did not exist at that time. In this contribution, we report about the effort undertaken at University of Liège to cautiously and consistently re-analyze and valorize these early observations, with modern tools.

The first steps to their re-analysis were to read them from old magnetic tapes, to take into account their zero offsets (measured before and after each recording), to carefully calibrate their wavenumber scale, to verify their associated airmass (approximate airmass routine was used at that time) and to save them in the format needed by the FTIR retrieval programs.

It is important to note that the non-linearity introduced when switching from the wavelength scale – original scale for a grating spectrum – to the wavenumber scale results in a negligible error, because of the small size of the recorded domains: for a micro-window of about 5 cm-1, relative total column error is not larger than 0.10 % for unsaturated lines.

Total columns have been derived with the retrieval algorithm SFIT-2 v3.91, which uses the optimal estimation method [4] to fit a synthetic spectrum to the observation. The pressure and temperature profiles adopted in the retrievals were provided by the National Centers for Environmental Prediction (NCEP, http://www.ncep.noaa.gov).

As the SFIT-2 software has been specifically developed for FTIR instruments with a sinc function as instrumental profile, we had to apply a strong Norton-Beer apodization [3] to the synthetic spectra to simulate at best the observed line shapes.

The total columns derived from grating spectra have been combined with the FTIR columns derived at the Jungfraujoch since the mid-1980s, in order to derive the temporal evolution of various target gases for the period 1977-2011. For water vapor, we hope to derive columns for a still longer period, thanks to systematic measurements of a water vapor line at 694.37 nm, initiated in 1974 to check the dryness of the atmosphere when recording solar atlases.

References

[1] Delbouille L. and Roland G., Photometric atlas of the solar spectrum from λ 7498 to λ12016. Mém. Soc. Roy. Sci. Liège, 1963, special vol 4.

[2] Delbouille L., Neven L. and Roland G., Photometric Atlas of the Solar Spectrum from λ3000 to λ10000. Institute of Astrophysics – Univ. of Liège, Special Vol., 1973–1988.

[3] Norton R. H. and R. Beer, “New Apodizing Functions for Fourier Spectrometry”, J. Opt. Soc. Am. 66, 259-264 (1976) and “Errata - New Apodizing Functions for Fourier Spectrometry”, J. Opt. Soc. Am., 67, 419 (1977).

[4] Rodgers, C.D., Inverse methods for atmospheric sounding: theory and practice, World Scientific, 2000.

[5] Zander, R., G. Roland, L. Delbouille, A. J. Sauval, C. B. Farmer and R. H. Norton, Monitoring of the integrated column of hydrogen fluoride above the Jungfraujoch station since 1977 - the HF/HCl column ratio, J. Atmos. Chem., 5, 385-394, 1987.

[6] Zander, R., E. Mahieu, P. Demoulin, P. Duchatelet, G. Roland, C. Servais, M. De Mazière, S. Reimann and C.P. Rinsland, Our changing atmosphere: Evidence based on long-term infrared solar observations at the Jungfraujoch since 1950, Sci. Total Environ., 391, 184-195, 2008. [http://hdl.handle.net/2268/2421]

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Oral Session

7-3 High precision ground based remote sensing observations of greenhouse gases within the Total Carbon Column Observing Network (TCCON)

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Throughout the last years the ground-based remote sensing has been further developed to measure the long lived greenhouse gases with high precision. Using the sun as light source FTIR measurements in the near infrared spectral region allow to retrieve averaged column mixing rations of CO2 and CH4 with a precision of < 0.3%. Currently the observations are performed worldwide at about 15 sites. At several sites the observations are performed using the NDACC FTIR instruments that have been upgraded to measure in the near infrared spectral region. The observations are organized in the Total Carbon Column Observing Network (TCCON). TCCON has been established in 2004 and has become a vital component in the global observing system for greenhouse gases. TCCON represents an indispensable validation resource for satellite measurements and adds important complementary information to the existing in situ measurements. The determination of the sources and sinks of greenhouse gases from atmospheric concentration measurements requires inverse modeling. Until recently these models were solely based on surface in-situ measurements. The confidence in the fluxes inferred from these in situ measurements is limited due the difficulties in describing the vertical transport in the model and due to the sparse spatial coverage of the sampling sites. Column measurements overcome some of the limitations of the in situ network. They are much less sensitive to vertical transport and therefore provide complementary information to the in situ measurements and space-borne sensors. Following the first observations by SCIAMACHY and AIRS the Japanese satellite GOSAT, dedicated for greenhouse gas observations, now allows to measure CO2 and CH4. We will describe the current status of the TCCON, show advances of the network calibration against the WMO standards, present studies towards a closer understanding of the carbon cycle and finally show current validation of satellite observations.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Oral Session

7-4 Overview over the GCOS Reference Upper Air Network (GRUAN)

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Records of upper air climate observations, be they from in-situ soundings, ground-based remote sensing instrumentation or satellites, are frequently limited in their usefulness to detect changes in climatically relevant atmospheric parameters owing to uncertainties arising from changes in instrumentation and practices. In particular atmospheric water vapor and atmospheric temperature, two of the most important essential upper air climate variables, have been plagued with numerous observational artifacts. It is virtually certain that similar challenges will pervade the future record from the global observing system and efforts need to be taken to maintain or even improve the ability to detect changes in atmospheric parameters. For over a decade the climate community has worked to instigate a reference quality network to ensure the future record, in particular for temperature and water vapor. These efforts have been taking shape as the GCOS Reference Upper Air Network (GRUAN), which in parts has adopted a number of lessons learned within the Network for the Detection of Atmospheric Composition Change (NDACC). This presentation will discuss the rationale behind GRUAN, progress thus far and future plans. A central element of GRUAN is the definition of a reference observation. It requires that measurements are traceable to an accepted standard, preferably an SI standard, that measurements are well documented and accompanied by extensive meta data, and that the level of confidence in these measurements is described by a vertically resolved uncertainty analysis. To verify that these estimates of measurement uncertainty remain within the expected limits redundant observations at regular intervals will be required. This verification is essential to maintain the current level of understanding of the measurement process and the subsequent processing. Furthermore it aides in identifying weaknesses in the observational process and provides guidance as to the remedy of these weaknesses. It is hoped that this observational strategy will provide tools to manage instrumental change, which has in the past often limited the ability to detect atmospheric variability and change. Not only does it allow quantitatively describing and correcting systematic differences in the response of different instruments, it also requires an increase in openness and transparency of the processing and analysis of the observations. Network homogeneity is an essential aspect of GRUAN and great efforts will be spent in assuring equal quality for equal measurements at different sites. In addition to frequent intercomparisons, central meta data management, long term archiving of raw data and central processing of identical instruments are essential elements required to achieve a network of similarly characterized and bias free observations over many decades.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Oral Session

7-5 Combining NDACC data with satellite observations to constrain hydrocarbons in chemistry-climate models

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The current generation of chemistry-climate models provides reasonable simulations of past variations in stratospheric circulation and constituents, as highlighted in the recent CCMval2 comparisons. One area for future improvement involves understanding and simulation of the coupled troposphere-stratosphere system, especially in models that include tropospheric hydrocarbons. We discuss comparisons of hydrocarbons observed by ground-based FTIR observations and space-based measurements (from the ACE-FTS instrument) with results from the NCAR Whole Atmosphere Community Climate Model (WACCM), based on climatological emissions and analyzed meteorological fields. Comparisons include aspects of the seasonal cycle, and longer-term variations derived from NDACC data.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Oral Session

7-6 Tropical tropopause layer transport and processing of short-lived substances

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The properties of the tropical tropopause layer have changed in recent decades, e.g. increases in greenhouse gases (GHGs), and associated changes in dynamics, have cooled the tropical tropopause layer thereby decreasing in situ water vapour and partially offsetting surface climate warming. From 1984 to 2005, ozone in the tropical tropopause layer declined, affecting radiative forcing in the tropical tropopause layer region. Water vapour in the tropical tropopause layer modulates much of the chemistry that affects ozone, sulfur and halogen compounds. As a result, understanding water vapour transport through the tropical tropopause layer, its chemical interactions with trace species, and associated aerosol and cloud formation, is a prerequisite to understanding the processing of sulfur and short-lived substances in the tropical tropopause layer. Here we present a detailed study of the dehydration processes through the tropical tropopause layer combined with convective influence using a microphysical model combined with Lagrangian transport to accurately capture the air parcel's temperature history. Aerosol formation via binary, ternary and ionic nucleation processes are included, ice formation via homogeneous and heterogeneous nucleation of existing aerosol, growth/evaporation (sublimation), coagulation, and sedimentation processes are included. Convective detrainment into the Lagrangian advected airmasses using ERA-Interim detrainment rates is also considered. The seasonality of the stratospheric water vapour at 400 K is reproduced and the sensitivity to convectively delivered sulfur species is discussed. The mechanism driving the increase in stratospheric sulfate aerosol in the recent volcanic quiescence period is currently not known. Contributions from increasing sulfur emissions, particularly over Asia and mid-sized volcanoes have been mooted. Within the model the delivery of sulfur gases to the tropical tropopause layer and stratosphere is studied. The aerosol formation and subsequent transport of convectively delivered short-lived sulfur species will be presented as a simple case study. Results from a conceptual study of very short lived bromine species which are currently considered to contribute between 3-8 ppt of stratospheric Bry (~25% of the total stratospheric Bry budget). The transport of these substances through the tropical tropopause layer is on a similar timescale to the lifetimes of these very short-lived compounds. Using a parameterised model run on Lagrangian back-trajectories we investigate the sensitivity of the delivered Bry to the various processes in the tropical tropopause layer transport. Sensitivity to sources, lifetimes, convective dilution of boundary layer air, and washout of inorganic bromine and the cold point temperature are considered.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Poster Session

7P-1 Strategy for high-accuracy-and-precision retrieval of atmospheric methane from the NDACC FTS network and intercalibration versus TCCON

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We present a strategy (MIR-GBM v1.0) for retrieval of column-averaged dry-air mole fractions of methane (XCH4) with a precision <0.3 per cent (1-sigma diurnal variation, 7-min integration) and a seasonal bias <0.14 per cent from mid-infrared ground-based solar FTIR measurements of the Network for the Detection of Atmospheric Composition Change (NDACC, comprising 22 FTIR stations). This makes NDACC methane data useful for satellite validation and for the inversion of regional-scale sources and sinks in addition to long-term trend analysis. Such retrievals complement the high accuracy and precision near-infrared observations of the younger Total Carbon Column Observing Network (TCCON) with time series dating back 15 years or so before TCCON operations began. MIR-GBM v1.0 is using HITRAN 2000 (including the 2001 update release) and 3 spectral micro windows (2613.70 - 2615.40 cm-1, 2835.50 - 2835.80 cm-1, 2921.00 - 2921.60 cm-1). A Tikhonov first derivative constraint is applied to the state vector given in units of per cent of volume mixing ratio. It is tuned to achieve minimum diurnal variation without damping seasonality. Final quality selection of the retrievals uses a threshold for the ratio of root-mean-square spectral residuals and information content (<0.15 per cent). Column-averaged dry-air mole fractions are calculated using the retrieved methane profiles and four-times-daily pressure-temperature-humidity profiles from National Center for Environmental Prediction (NCEP) interpolated to the time of measurement. MIR-GBM v1.0 is the optimum of 24 tested retrieval strategies (8 different spectral micro-window selections, 3 spectroscopic line lists: HITRAN 2000, 2004, 2008). Dominant errors of the non-optimum retrieval strategies are HDO/H2O-CH4 interference errors (seasonal bias up to 4 per cent). Therefore interference errors have been quantified at 3 test sites covering clear-sky integrated water vapor levels representative for all NDACC sites (Wollongong maximum = 44.9 mm, Garmisch mean = 14.9 mm, Zugspitze minimum = 0.2 mm). The same quality ranking of the 24 strategies was found for all 3 test sites with one optimum, i.e., MIR-GBM v1.0. Seasonality of XCH4 above the Zugspitze (47 N) shows a minus-sine shape with a minimum in March/April, a maximum in September, and an amplitude of 16.3+/-2.9 ppb (1.0+/-0.2 per cent). This agrees very well with newest-version scientific retrievals from SCIAMACHY. A conclusion from this work is that improved spectroscopic parameters for CH4, HDO, and H2O in the 2613 – 2922 cm-1 spectral domain are urgently needed. If such become available with sufficient accuracy, at least two more spectral micro windows could be utilized leading to another improvement in precision. NDACC-TCCON comparisons are performed using data from stations with coincident mid-IR (NDACC) and near-IR (TCCON) measurements (Ny-Alesund, 78.9 N, Bremen, 53.1 N, Karlsruhe, 49.1 N, Garmisch, 47.5 N, Izana, 28.3 N, Wollongong, 34.5 S, Eureka, 80 N). The overall goal of this study is to quantify the NDACC-

TCCON intercalibration. One question to be addressed is whether or not one overall intercalibration factor for all stations can be found. Furthermore, we have investigated whether the time series of intercalibration factors show a significant seasonal or latitudinal component which can arise due to airmass-dependent artifacts or HDO/H2O interference errors and could be parameterized.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Poster Session

7P-2 The vertical profiles of CH4 observed at Tsukuba with a Fourier transform spectrometer

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Fourier transform spectrometer (FTS) has advantages in its high resolution and the wide wavenumber range. Vertical profiles of some species can be derived from the high-resolution spectra. The vertical profiles and column densities of CH4 were retrieved from the solar spectra observed at Tsukuba, Japan. CH4 is relatively stable and second important greenhouse gas. But source intensities and their temporal variations aren't understood well. Therefore, the mechanism of the variation of increasing rate of surface concentration of CH4 isn't understood too. In the stratosphere, CH4 is only destructed and finally produce H2O. There aren't many observations of temporal variations of stratospheric CH4 and quantitative understanding isn't sufficient. Observation at Tsukuba started in 1998 with Bruker 120M. 120HR was used from 2001 and is switched to 125HR from 2010. The resolution of the spectrum is 0.0035 cm-1. The retrieval of vertical profiles is performed using the spectra taken with 120HR and 125HR because the optics of our 120M wasn't so good. The vertical profiles of CH4 ware retrieved with SFIT2 spectral fitting program developed by Rinsland et al. (1998). In the analysis, we have to select an appropriate wavenumber region and the optimization of fitting parameters are also needed. Now we used 2904 cm-1 region for preliminary analysis. NDACC/IRWG group is investigating the optimization of microwindows and the parameters and we will reanalyze the vertical profiles and column densities of CH4. We found that the phase of seasonal variation of the mixing ratios in the lower stratosphere is shifted from those in the troposphere. The maximum occurred in autumn and the minimum occurred in spring in the stratosphere due to circulation but the maximum occurred in winter and minimum occurred in summer in troposphere due to reaction with OH. The temporal variation of total column shows step-like increase in 2007 as reported by other observations.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Poster Session

7P-3 Spectroscopic Study of CH4 and CO Total Column Amount in the Atmosphere near Saint-Petersburg, Russia – Maria Makarova

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Methane is one of the most important "greenhouse" gases in the Earth's atmosphere. Currently, the relative contribution of CH_4 to radiation forcing is of about 20% [1]. plays a noticeable role in the atmospheric chemistry: in the troposphere, its reactions with OH and O_3 are important; in the stratosphere, methane is a source of H_2O and hydrogen and a sink of chlorine [2]. Carbon monoxide significance in atmospheric chemistry lies mainly i its competition with many other gaseous pollutants—importantly the greenhouse gas CH_4—for the hydroxyl radical (OH, $CO + OH \rightarrow CO_2 + H$). Increased CO emissions cause higher CO burdens and more reaction with OH, leaving less OH for cleansing the troposphere of other reduced gases.

Spectroscopic measurements of atmospheric CH_4 total column amounts (TCAs) (1991-2011) and CO TCAs (1995-2011) are carried out by Dept. of Atmospheric Physics, Research Institute of Physics, Saint-Petersburg State University [3]. Measurement site is situated in Peterhof (suburb of Saint-Petersburg, about 35 km to the west from the megacity center), 59.88N, 29.83E, 20m asl. Grating IR solar spectrometer (SIRS) with spectral resolution of 0.5 cm^{-1} is in operation from 1991. Fourier spectrometer Bruker IFS125 HR (spectral resolution is up to 0.002 cm-1) started observations of IR solar radiation in January 2009 (Fig.2). Original sun-tracker system for Bruker IFS125 HR was designed at the Dept. of Atmospheric Physics of Saint-Petersburg State University (Fig.3, 4).

Fig. 1 Fourier spectrometer Bruker IFS125 HR.

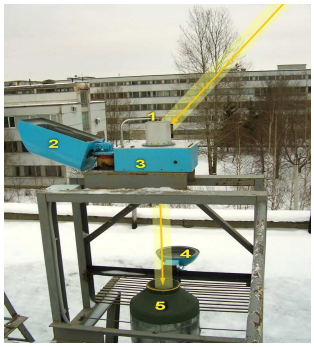

Fig. 2 Original sun-tracker system (on the roof) for Bruker IFS125 HR designed at the Dept. of Atmospheric Physics.

Fig. 3 Original sun-tracker system for Bruker IFS125 HR designed at the Dept. of Atmospheric Physics.

Simultaneous measurements which have been performed by SIRS (original retrieval algorithm [4]) and Bruker (SFIT2 retrieval algorithm [5]) during 2009-2010 allow us to

harmonize CH_4 and CO time series obtained by two spectrometers. For example, systematic difference between Bruker IFS125 HR and SIRS measurements was of 1.8% (Fig. 4). Harmonized CH4 TCA time series for 2009-2010 (systematic difference between Bruker IFS125 HR and SIRS measurements was taken into account) are illustrated in Fig.5.

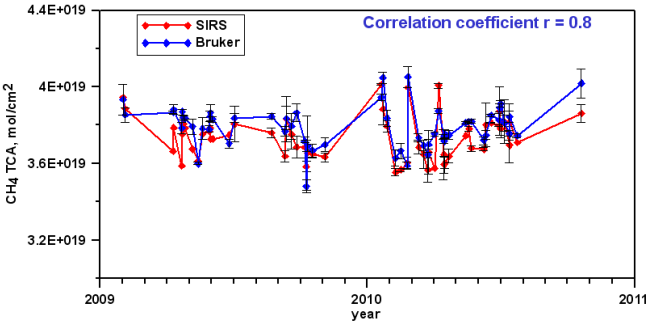

Fig.4 Results of simultaneous measurements of CH4 TCA.

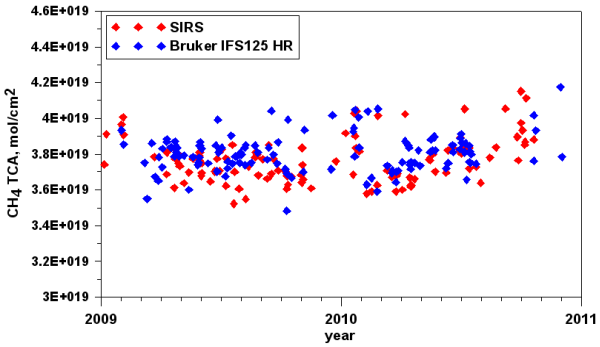

Fig. 5 Harmonized CH4 TCA time series for 2009-2010.

Growth rates, seasonal cycles and interannual variability of CH_4 and CO TCAs are estimated for the measurement periods using data of both spectrometers. CH_4 TCA has no statistically significant trend for the period 1991-2005 – (- 0.06 ± 0.2)% per year, however from 2006 CH_4 TCA started to grow (Fig.6). Increase rate of CH_4 TCA for 2005-2010 is of (1.0 ± 0.4) % per year (Fig.6). Long-term linear trend of CH4 (near Saint-Petersburg) for all measurement period (1991-2010) amounts (0.16 ± 0.1)% per year (Fig.6).

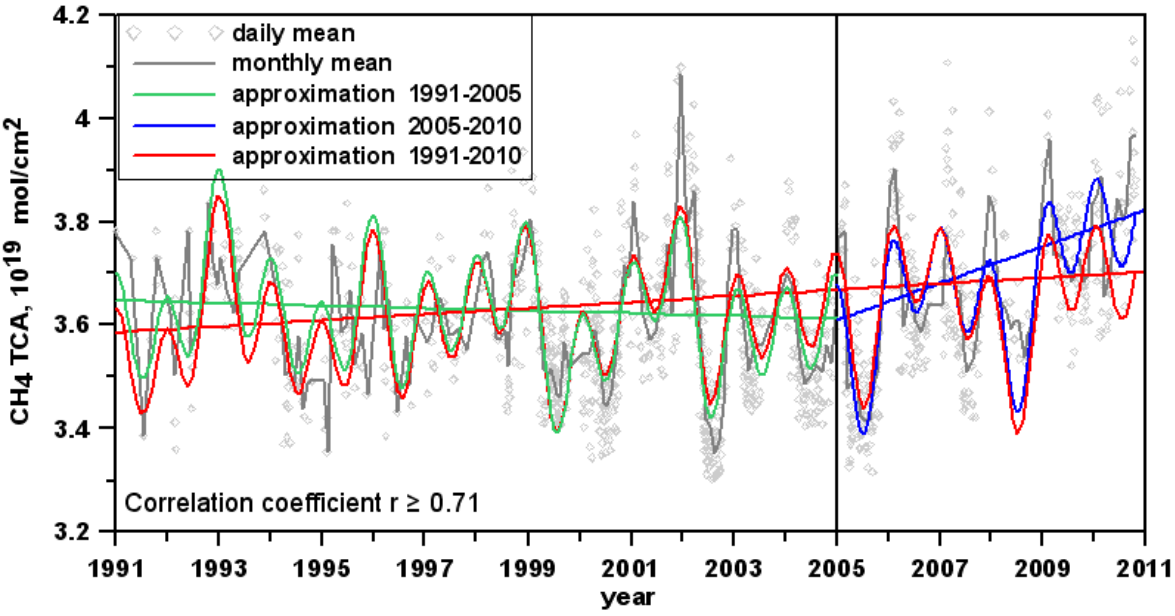

Fig.6 Long-term trends of CH4 TCA near Saint-Petersburg.

CO TCA measurements for 1995-2010 are presented in Fig. 7. It was shown that the irregular component of the CO TCA (CO anomalies, calculated for the period from May to September) is in good agreement with data on the area of forest fires and the volume of the burnt wood (Fig. 8). According to our analysis the fewest number of forest fires were observed in 1999, 2001, 2005, 2007 and 2009.

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REFERENCES
1. IPCC Fourth Assessment Report (AR4) “Climate Change 2007”, Working Group I Report “The Physical Science Basis,” http://www.ipcc.ch/pdf/assessment-report/ ar4/wg1/ar4-wg1-chapter2.pdf.
2. Scales and Global Changes: Spatial and Temporal Variability in Biospheric and Geospheric Processes, Ed. by T. Rosswall, R.G. Woodmansee, and P.G. Risser, (Scientific Committee on Problems of Environment, Wiley, 1988).
3. Makarova M. V., A. V. Poberovskii, K. N. Visheratin, and A. V. Polyakov. Time Variability of the Total Methane Content in the Atmosphere over the Vicinity of St. Petersburg // Izvestiya, Atmospheric and Oceanic Physics, 2009, Vol. 45, No. 6, pp. 723–730.
4. A. V. Mironenkov, A. V. Poberovskii, and Yu. M. Timofeev. Interpretation of Infrared Solar Spectra for Quantification of the Column Content of Atmospheric Gases. Izv., Atmos. Ocean. Phys. 36 (2), 191–198 (1996).
5. Rinsland, C. P., Jones, N. B., Connor, B. J., Logan, J. A., Pougatchev, N. S., Goldman, A., Murcray, F. J., Stephen, T. M., Pine, A. S., Zander, R., Mahieu, E., and Demoulin, P.: North- 20 ern and southern hemisphere ground-based infrared spectroscopic measurements of tropospheric carbon monoxide and ethane, J. Geophys. Res.-Atmos., 103, 28197–28217, 1998.
6. Byun, D.W. and K. Schere. Review of the Governing Equations, Computational Algorithms, and Other components of the Models-3 Community Multiscale Air Quality (CMAQ) Modeling System. Applied Mechanics Review. 2006. 59, 51-77.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Poster Session

7P-4 Changes in atmospheric composition discerned from long-term NDACC measurements: Tropospheric gases measured by infrared Fourier transform spectroscopy at Thule, Greenland

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Broadband high-resolution infrared solar spectra contain information on many species of importance to stratospheric and tropospheric chemistry and global warming are obtained from the IRWG FTS instruments. Advances in retrieval methods allow the vertically resolved discrimination of many constituents. It has long been known that lower latitude air masses often containing high concentrations of anthropogenic or biomass burning pollutants are transported into the Arctic. Here they may accumulate, especially during the Arctic night, when photolysis and oxidation are minimized. Though changes to a warmer Arctic its expected that more local biogenic emissions will contribute to tropospheric chemical activity. The high Arctic NDACC IRWG site at Thule, Greenland (76.5N, 68.7W, 225masl) has been in continuous operation since 1999 making observations typically from late February through September during times excluding the polar night. Total column amounts and low-resolution vertical mixing ratio profiles of a number of gases important to tropospheric chemistry and to greenhouse warming are analyzed and their evolution characterized. Annual cycles and long-term trends are shown for CH4, N2O, CO, C2H6, C2H2, HCN and HCOOH. Specific mid year pollution events are identified as perturbations of the background state. Upper limits are assigned to several other important gases that are near or below current detection limits.

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Poster Session

7P-5 Long-term trends of a dozen direct greenhouse gases derived from infrared solar absorption spectra recorded at the Jungfraujoch station

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The University of Liège (ULg) is operating –under clear sky conditions– two state-of-the-art Fourier Transform Infrared (FTIR) spectrometers at the high-altitude research station of the Jungfraujoch (Swiss Alps, 46.5ºN, 3580m asl), within the framework of the Network for the Detection of Atmospheric Composition Changes (NDACC, http://www.ndacc.org). Routine FTIR operation started in 1984. Since then, it has been continued without disruption, allowing collecting more than 45000 high-resolution broadband infrared solar absorption spectra, between 2 and 16 microns, using either HgCdTe or InSb detectors as well as a suite of optical filters. Typically, the spectral resolutions achieved lie in the 0.003 to 0.009 cm-1 interval while signal-to-noise ratios of 1000 and more are reached. In addition, numerous narrow-band IR spectra essentially recorded from 1976 to 1989 with grating instruments are also available [e.g. Zander et al., 2008]. Their analyses with modern tools have recently started [Bader et al., 2011] and will be pursued in the coming years to consistently extend our total column datasets back in the 1970s.

Geophysical parameters are deduced from the ULg observational database either with the SFIT-1, the SFIT-2 or the PROFFIT-9 algorithms, allowing producing long-term total column time series of the target gases. In addition, information on their vertical distributions with altitude can generally be derived when using SFIT-2 or PROFFIT-9 which both implement the Optimal Estimation Method of Rodgers [1990]. Presently, more than two dozen atmospheric species are systematically retrieved from the Jungfraujoch observations, allowing the monitoring of key constituents of the Earth's atmosphere which play important roles in global warming and/or in stratospheric ozone depletion.

The present communication will focus on the direct and major greenhouse gases available from our database, namely water vapor, CO2, CH4, N2O, tropospheric ozone, CFC-11, CFC-12,

HCFC-22, CCl4, SF6, as well as CF4 which has recently been added to our targets list [Duchatelet et al., 2011]. Trends and associated uncertainties characterizing the available –and often multi-decadal– time series have been derived or updated with a statistical bootstrap resampling tool [Gardiner et al., 2008], they will be presented and critically compared with data available from the literature.

Acknowledgments

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This communication is dedicated to our colleague and friend Curtis P. Rinsland who passed away in April 2011. We have enjoyed and benefited from a close and steady collaboration with him, over nearly 30 years for some of us, within the framework of the ATMOS, ACE and NDACC programs and missions. We will deeply miss him

References

Bader, W. et al., 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, *Geophysical Research Abstracts, 13*, EGU2011-3391-1, 2011. [http://hdl.handle.net/2268/88180]

Duchatelet, P. et al., First retrievals of carbon tetrafluoride (CF_4) from ground-based FTIR measurements: production and analysis of the two-decadal time series above the Jungfraujoch, *Geophysical Research Abstracts, 13*, EGU2011-6413, 2011. [http://hdl.handle.net/2268/90745]

Gardiner, T. et al., Trend analysis of greenhouse gases over Europe measured by a network of ground-based remote FTIR instruments, *Atmos. Chem. Phys., 8*, 6719-6727, 2008. [http://hdl.handle.net/2268/2545]

Rodgers, C.D., Characterisation and error analysis of profiles derived from remote sensing measurements, *J. Geophys. Res.*, 95, 5587-5595, 1990.

Zander, R. et al., Our changing atmosphere: Evidence based on long-term infrared solar observations at the Jungfraujoch since 1950, *Sci. Total Environ., 391*, 184-195, 2008. [http://hdl.handle.net/2268/2

2011 NDACC Symposium
Network for the Detection of Atmospheric Composition Change

Poster Session

7P-6 Observation of Middle Atmosphere in Río Gallegos NDACC site, Argentina

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Since spring 2005 the Lidar Division of CEILAP, joined with French and Japanese researchers has been monitoring atmospheric parameters using lidar technologies in southern most continental part of South America. The Río Gallegos experimental site is located in South Patagonia (51º 55'S, 69º 14'W), in subpolar region and it is a convenient monitoring site of the atmosphere in the southern hemisphere. This experimental site operates a differential absorption lidar instrument (DIAL) for the measurement of ozone vertical distribution. This instrument is part of Network Data for Atmospheric Composition Change (NDACC). The altitude range of the ozone measurement is 14-45 km, which provides the opportunity to monitor the perturbations due to the passage of stratospheric polar air over Río Gallegos. Systematic stratospheric ozone profile measurement has been carried on in this experimental site since 2005. We identified three mayor perturbation of ozone hole over the stratospheric ozone profile in Río Gallegos. Approach of polar vortex during late winter, overpass of ozone hole in middle spring and dilution process during late spring change the shape and content of stratospheric ozone profile and by consequences the solar UV. In 2006 we began to obtain other products such as aerosol content and temperature profile in the stratosphere. The determination of temperature measurements from the Rayleigh scattering is an important remote sensing technique for obtaining stratospheric profiles. In this paper we showed the technique to measure temperature profiles in the stratosphere between 15-60 km altitude. The inversion temperature from photoncounting is detected from light scattered by the Rayleigh line at 355 nm generated from a laser Quantel YG-980. An analysis of the errors involved in the inversion process and analysis of a case of study is showed for the period 2005-2007. The results presented in this paper are validated through intercomparisons with measurements made by HIRDLS instrument (High Resolution Dynamics Limb Sounder) onboard the NASA AURA satellite platform, radiosounding and NCEP database. Currently the site is part of the UVO3Patagonia in collaboration with the laboratory of Ozone and UV Radiation in the city of Punta Arenas, Chile distant 200 km.

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Poster Session

7P-7 Rayleigh LIDAR investigation of major and minor sudden stratospheric warming observed over Northern hemisphere stations

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The stratospheric sudden warming is the most dramatic phenomenon to be observed in winter stratosphere over the globe. According to WMO definition, 'during some winters, zonal-mean configuration is dramatically disrupted with polar stratospheric temperatures increasing rapidly with time, leading to a poleward increase of zonal-mean temperature and, on occasion, a reversal of zonal-mean winds to an easterly direction exists. Such an event is called as Stratospheric Sudden Warming' (here after referred to as SSW). Since the first observation by Scherhag (1952), there have been large numbers of evidence of SSW in Northern and Southern hemisphere. Most of these warming are attributed to planetary wave breaking. So far, the SSW study is addressed to high, mid latitudes stations using limited datasets. Also, most of the results are based on satellite data, which are less in accuracy in comparison to the ground based measurements (like LIDARs). Recently, Sivakumar et al. (2004) has shown a first finding of stratospheric warming over tropics, and illustrated that the warming is due to increase in planetary-wave activity. Their calculation on Eliassen-Palm (E-P) flux from ECMWF reanalysis show clear evidence of propagation of planetary-wave activity from high and mid- to low latitudes consecutive to the major warming episode over the pole. The study evidenced that the SSW is not only focused to high/mid latitudes, it can extend to low latitude depend on the strength of warming and by meridianal circulation. Based on their study, we aimed to study the SSW event over three Northern hemisphere stations using the ground based LIDAR measurements and large data sets. Mainly, we focused our studies using the LIDAR data over tropics and mid-latitude stations, i.e., Gadanki (13.5ºN; 79.2ºE), Mauna Loa (19.5ºN; 155.6ºE) and OHP (44ºN; 6ºE). Two of the above LIDAR stations (Mauna Loa and OHP) are in operational under Network Detection for Atmospheric Composition Change (NDACC). We use the Rayleigh LIDAR temperature data of 20 years (1981-2001) over OHP, 18

years (1994-2005) over Mauna Loa and 6 years (1998-2004) over Gadanki sites. The warming events are noticed in the data sets and are classified into major and minor warmings based on the temperature increase and reversal in the zonal wind direction in the polar region using National Centre for Environmental Prediction (NCEP) reanalysis data. The statistical characteristics of warming events are obtained and reviewed based on the magnitude of warming, height of occurrence, day and period of occurrence with special emphasis on wave propagation and isentropic transport conditions. Reference: Sivakumar,V., B. Morel, H. Bencherif, J. L. Baray, S. Baldy, A. Hauchecorne., and Rao.P.B.: Rayleigh lidar observation of a warm stratopause over a tropical site, Gadanki (13.5℃N; 79.2℃E), Atmos. Che m. Phys., 4, 1989--1996, 2004.