

# AN OVERVIEW OF NDSC-RELATED ACTIVITIES AT THE JUNGFRAUJOCH THROUGH HIGH-RESOLUTION INFRARED SOLAR OBSERVATIONS

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**Abstract.** Since the mid-1970s, infrared remote sensing at the Jungfraujoch, Switzerland, has been performed in response to the need to measure the concentrations and assess the seasonal and long-term changes of an increasing number of atmospheric constituents. Currently, the vertical column abundances of over 20 telluric gases are being monitored, based on the analysis of solar spectra recorded regularly between 2 and 14  $\mu\text{m}$ , using two high-resolution Fourier transform spectrometers. In this paper, emphasis is placed on those activities more specifically performed within the frame of the NDSC (Network for the Detection of Stratospheric Change) whose objectives are briefly described in the introduction.

## Introduction

Since the early 1970s, numerous field measurements have revealed series of changes in the composition of the Earth's atmosphere, mainly under man-made influences, resulting primarily from increasing energy consumption and from modern industrial and agricultural practices. This anthropogenic impact has often been offered as a threat to the mean living conditions on Earth, from both climatic and biological points of view. One of the serious concerns resulted from the accumulation in the troposphere of a series of new and widely used halogenated source gases, primarily the chlorofluorocarbons CFC-11 and CFC-12: their long lifetimes would allow them to reach the stratosphere, photodissociate there, and threaten the protective ozone layer by releasing their chlorine atoms which activate the  $\text{ClO}_x$  catalytic cycle [Molina and Rowland, 1974]. Confirmation of this hypothesis was the detection of the main chlorine reservoir HCl in the stratosphere in 1975 [Farmer et al., 1976; Ackerman et al., 1976]. The fluorine reservoir HF had already been detected in the upper stratosphere in 1974 [Zander, 1975], a unequivocal confirmation that CFCs are indeed photodissociated above 25 km altitude (as no natural fluorinated source gas could explain the presence of HF at such altitudes). Soon after, HCl and HF were detected then monitored through the analysis of ground-based infrared solar spectra recorded at the International Scientific Station of the Jungfraujoch (ISSJ), Switzerland [Zander et al., 1977, 1987a, 1987b]; this was the beginning of an atmospheric monitoring effort by the Liège group, initially supported by the CMA (Chemical Manufacturers Association, Washington, DC, USA).

In response to the above and many other changes observed till the mid-1980s [see WMO-Report No. 16, 1986], in particular the steady increase of the inorganic chlorine loading in the stratosphere [e.g., Mankin and Coffey, 1983; Zander et al., 1987b] and the dramatic spring time reduction of ozone discovered over Antarctica during the early 1980s [Farman et al., 1985], a group of experts met in Boulder, CO, on March 5-7, 1986, to evaluate the means of measuring such changes

on a global and consistent basis and to identify their causes. It led to the inception of the "Network for the Detection of Stratospheric Change" (NDSC).

The NDSC is a set of ground-based atmospheric research stations equipped with high-quality, remote-sounding instruments to monitor the physical state and the chemical composition of the earth's middle atmosphere at various latitudes in both hemispheres. Emphasis is placed on long-term observations through which changes in the stratosphere can be determined and understood, in particular those affecting the ozone layer.

The NDSC went into official operation in 1991 [Kurylo and Solomon, 1990]. It is articulated on both "primary" and "complementary" sites. Presently, there are five primary stations, i.e., Arctic (Eureka, NyÅlesund, Thule), Alpine (Jungfraujoch, Plateau de Bure, Haute Provence), Hawaiian (Mauna Loa, Mauna Kea), New Zealand (Lauder) and Antarctic (Dumont d'Urville, McMurdo, Dôme Concorde), spanning 80° N to 78° S. Over two dozen complementary sites are contributing limited but quality-controlled scientific input of relevance to the network, further broadening its international status and geographical coverage.

Among the instruments selected for NDSC-priority measurements (on the basis of being remote sensors capable of long-term field operation with proven performances and stability; i.e., UV-Vis-,  $\mu\text{wave}$ - and Fourier transform infrared spectrometers, and lidars), the Fourier transform infra-red spectrometers were initially tasked to monitor the vertical column abundances of HCl,  $\text{ClONO}_2$ ,  $\text{HNO}_3$ ,  $\text{CH}_4$ , and  $\text{N}_2\text{O}$ , with an additional suite of species ( $\text{H}_2\text{O}$ ,  $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{O}_3$ ,  $\text{NO}$ ,  $\text{NO}_2$ ,  $\text{C}_2\text{H}_2$ ,  $\text{C}_2\text{H}_6$ ,  $\text{HCN}$ ,  $\text{OCS}$ , CFC-11, CFC-12, HCFC-22,  $\text{CCl}_4$ ,  $\text{COF}_2$ , HF,  $\text{SF}_6$ ,...) to be investigated when time permits.

Since 1990, the long-term funding of the European involvement at NDSC-primary stations has been partly ensured through successive ESMOS (European Stratospheric Monitoring Station) programs [Simon et al., 1990].

In the following paragraphs, we present and briefly comment typical column abundance data bases derived from FTIR solar observations made at ISSJ within the frame of the infrared component of the NDSC primary Alpine station. We further evoke instrumental and analytical intercomparison activities which have been conducted over the last years.

## ISSJ Observations and Data Analysis

The ISSJ is located in the Swiss Alps, near Interlaken, at 3580 m altitude (latit. 46.55° N; longit. 7.98° E). Since 1990, the Liège group, in collaboration with colleagues from the Belgian Institute for Space Aeronomy (BISA), operates two high resolution Fourier transform spectrometers (one home-made prototype that was installed in 1984 and a Bruker IFS-120 HR acquired in 1990, achieving 0.0025 and 0.001  $\text{cm}^{-1}$

resolution, respectively) which can be operated between 2 and 14  $\mu\text{m}$  using  $\text{CaF}_2$ ,  $\text{ZnSe}$  and  $\text{KBr}$  beamsplitters, and cooled  $\text{InSb}$  and  $\text{HgCdTe}$  detectors. Prior to the FTIR instruments, the station was equipped with high performance grating spectrometers, primarily used for solar studies in the visible, near- and middle infrared [Migeotte et al., 1956; Delbouille et al., 1988]; during regular short time periods in 1950-51 and after 1976, they were also operated to support atmospheric investigations. Further details about the ISSJ instrumentation can be found in Delbouille and Roland [1995].

Over thirty telluric constituents possess characteristic absorptions in the 2 to 14  $\mu\text{m}$  interval [see Table 1 in Brown et al., 1992], with most of their spectroscopic line parameters being sufficiently well known so that column abundances for most of them can be evaluated to better than  $\pm 5\%$ , one sigma [Brown et al., 1996]; one important exception is  $\text{ClONO}_2$  whose 1-sigma retrieval accuracy remains still in the 10 to 15% uncertainty range. Notice that column variability and long-term trend investigations are primarily affected by the quality and internal consistency of the observations as well as the current uncertainty in the "reference" volume mixing ratio profiles adopted along the spectral fitting process, little by line parameters uncertainty. The vertical column abundances of the target gases are derived through the nonlinear least-squares (NLLS) spectral fitting procedure between observed and calculated spectra; the latter use some a priori, "most realistic" volume mixing ratio (VMR) profiles for both target and interfering gases which are iteratively scaled uniformly over their entire altitude range until the residuals of the fit reach a minimum. The two major and competing sources of uncertainty in this approach are the instrumental line shape (ILS) and the a priori VMR profiles adopted; this was clearly identified along retrieval algorithm intercomparison exercises carried out as part of ESMOS/NDSC-related quality assessment activities [Zander et al., 1993]. Unless the ILS of the FTIR instrument used to make the observations is properly characterised, it will be impossible to reliably progress in deriving altitude information on the current VMR profile from the NLLS spectral fitting of ground-based observations.

### Typical ESMOS/NDSC-Related Results

Figure 1 reproduces a subset of vertical column abundance measurements for 8 target molecules routinely monitored since 1985, and specifically as part of the ESMOS/NDSC-related activities since 1990; the points correspond to daily mean vertical columns expressed in number molecules per  $\text{cm}^2$  above the ISSJ (notice the different vertical scales and exponents to the base ten for the various gases).

The molecules  $\text{N}_2\text{O}$  and HF displayed in Frames A and H are included as tracers of dynamics. While the former has its sources at the ground and displays a quasi-constant VMR distribution through the troposphere, with a marked decrease above the tropopause, the latter is primarily concentrated in the stratosphere as a result of photodissociation of fluorinated sources of anthropogenic origin (e.g., CFCs and HCFCs). Consequently, the  $\text{N}_2\text{O}$  and HF column variations are anti-correlated when vertical dynamic motions occur; this is observed, indeed, all along the displayed data base in Fig. 1. Most clearly noticed are the variations during the winter months of 1991-92 and 1992-93, when significant masses of polar air intruded over the European continent; these extreme variations superimpose on a seasonal modulation which is

also anti-correlated for  $\text{N}_2\text{O}$  and HF, the latter reflecting tropopause height changes during the year.

The other six molecules in Fig. 1, i.e.,  $\text{O}_3$ ,  $\text{NO}_2$ ,  $\text{NO}$ ,  $\text{HNO}_3$ ,  $\text{ClONO}_2$  and  $\text{HCl}$  are all of stratospheric origin and their monitoring is, therefore, fully in line with the ESMOS/NDSC main objectives described before. Characteristic key features for each of them as derived from the vertical column data bases gathered between the mid-1980s and 1995 will be pointed out hereafter.

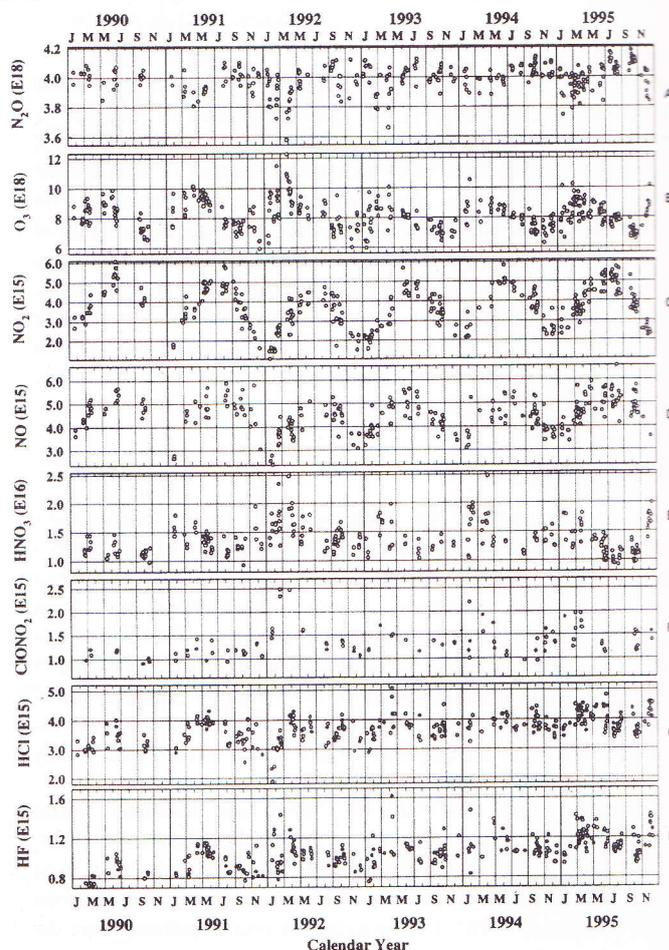


Figure 1. Sample vertical column abundances above ISSJ of eight atmospheric constituents primarily measured within the frame of ESMOS/NDSC stratospheric monitoring activities.  $\text{N}_2\text{O}$  and HF are included as tracers of dynamics.

### Ozone ( $\text{O}_3$ )

$\text{O}_3$  columns undergo a well defined seasonal variation with an average peak-to-peak amplitude of 25%, the maximum occurring in March and the minimum in October-November. The entire 1984-1995 data base reveals a long-term decrease of  $(7.0 \pm 1.5)\%$  per decade; when excluding the data points for the fraction 0.0 to 0.3 of each year (i.e., prior to April 20) during which the column variability is relatively large, one finds a rate of decrease which is only  $(3.5 \pm 1.0)\%$  per decade. This timely differentiation thus indicates that the decrease of the ozone burden above Europe during the last decade was substantially larger in winter and early spring than during the rest of the year. Similar findings have been reported by Bojkov et al. [1995], based on northern midlatitude Dobson

measurements. Thus far, chemical-dynamical model studies have been unable to properly reproduce, neither this ozone decrease, nor its time-differential change. The approach for an improved evaluation of total ozone including FTIR and UV-vis SAOZ (Système d'Analyse par Observations Zénithales) observations made at ISSJ, is reported elsewhere in these proceedings [De Mazière et al., 1997a]. Notice the relatively lower O<sub>3</sub> columns from the winter 1991-92 to mid-1994. This decrease has been attributed to the increased stratospheric sulphuric aerosol loading resulting from the volcanic eruption of Mt. Pinatubo (Philippines) in June 1991; the excess loading led to the conversion of N<sub>2</sub>O<sub>5</sub> into HNO<sub>3</sub> [Rinsland et al., 1994], with the immediate result that NO<sub>x</sub> decreased and that higher levels of active chlorine became available to destroy O<sub>3</sub>.

### Nitrogen compounds (NO<sub>2</sub>, NO, HNO<sub>3</sub>, ClONO<sub>2</sub>)

The odd nitrogen chemistry in the stratosphere is critical, because of its ability to destroy ozone directly through catalytical NO<sub>x</sub> reactions and indirectly by affecting the partitioning of odd hydrogen and odd chlorine species [Crutzen, 1971; Molina and Rowland, 1974]. Its complex assimilation in CTM models, resulting from the latter family interactions, dynamical phenomena and seasonal changes, has improved substantially [Crutzen et al., 1995], but significant discrepancies persist [e.g. Prather and Remsberg, 1993; Newchurch et al., 1996].

Based on the 1985 to 1995 ISSJ data bases, observational findings for the nitrogen compounds dealt with here can be summarised as follows:

	NO <sub>2</sub>	NO	HNO <sub>3</sub>	ClONO <sub>2</sub> <sup>(1)</sup>
Peak-to peak ampl. (%)	80	32	33	--
Time of max. ampl.	Jun-Jul	Jun-Jul	Feb-Mar	--
Time of min. ampl.	Jan	Jan	Aug	--
1985-95 trend (%/yr)	+ 0.4	insignificant	insignificant <sup>(2)</sup>	+ 4.5
Mean 1990 column	3.60 x E15	4.50 x E15	1.36 x E 16	1.10 x E15

<sup>(1)</sup> for observational and analytical details, see Rinsland et al. [1996]

<sup>(2)</sup> confirming earlier investigation by Rinsland et al. [1991]

On average, the NO<sub>y</sub> budget (limited here to the summation of the daytime columns of NO<sub>2</sub> + NO + HNO<sub>3</sub> + ClONO<sub>2</sub>) increased by (0.30 ± 0.06) %/yr, which is in excellent agreement with the (0.31 ± 0.05) %/yr rate of change observed above ISSJ for the main source gas N<sub>2</sub>O during the same time. The NO<sub>2</sub> timeseries from FTIR has been combined with that from the co-located SAOZ that comprises daily morning and evening vertical column amounts derived from measurements in the 87° to 91° solar zenith angle range. The mutual agreement between both datasets and the methodology for combination taking into account the NO<sub>2</sub> diurnal variation are described by De Mazière et al. [1997b]. The combined NO<sub>2</sub> timeseries has allowed a precise quantitative evaluation of the impact of the Mt. Pinatubo eruption on the NO<sub>2</sub> column abundance above ISSJ: the reduction was largest in Dec. 1991- Jan. 1992, amounting to 40% with respect to the 1988-90 mean level, and recovered to the latter base level by the end of 1994, with an exponential E-folding time of about 20 months. Both NO<sub>2</sub> and NO decreases resulted from the heterogeneous conversion of N<sub>2</sub>O<sub>5</sub> to HNO<sub>3</sub> on the surfaces of sulphuric aerosols.

### Inorganic chlorine loading (HCl and ClONO<sub>2</sub>)

While HCl and ClONO<sub>2</sub>, the two most important reservoirs of inorganic chlorine in the stratosphere can be monitored routinely from the ground, the less abundant ClO radical cannot at its mid-latitude background level. Frame G of Fig. 1 reproduces column abundances of HCl above ISSJ between 1990 and 1995. Notice the very low HCl columns observed along with large HF values in January 1992, indicating that processed masses of air from inside the polar vortex propagated over Europe; the airmasses sounded in April 1993 contained large amounts of both HCl and HF, resulting again from polar air intrusions, but without HCl depletion by heterogeneous processes. Findings derived from the longer 1976-95 HCl data base include (i) a mean exponential rate of increase of (4.1 ± 0.2) %/yr; (ii) a seasonal variation with a peak-to-peak amplitude of about 12 % of the local column, with the maximum occurring in April-May and the minimum in Sept.-Oct.; (iii) significant short-term variability during winter-spring time, primarily related to meridional transport.

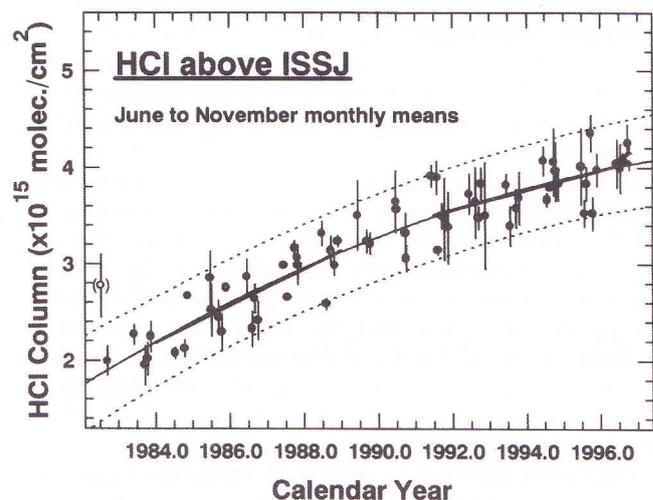


Figure 2. Monthly mean vertical column abundances of HCl above ISSJ for the "quiet" months June to November from 1982 to present. The 2nd order polynomial fit to the filled data points indicates a definite slowing down of the rate of increase of HCl above the site. The high column observed in July 1982 (open circle) is presumed to contain a bias caused by the El Chichon volcanic eruption of March-April 1982 [Mankin and Coffey, 1983].

To better quantify the HCl rate of change which is sensitive to both seasonal and short-term variations, we have reproduced in Figure 2 the monthly mean HCl columns for the "quieter" months of June to November available from 1982 to 1996. Fitting a 2nd order polynomial function to all filled data points of Fig. 1 (continuous thin curve) and the 95 % confidence limits (dotted curves) indicates that the rate of increase of the HCl column above ISSJ has slowed significantly during the last years. Linear fits to 5 year periods spanning from 1984.0 to 1989.0 and from 1991.5 to 1996.5 (see the thicker lines in Fig. 2) overlap very well with the 2nd order fit, thus indicating internal consistency of the data base. The rates of HCl increase derived for the two periods considered above are equal to 0.19 ± 0.02 and 0.10 ± 0.02 x 10<sup>15</sup> molec./cm<sup>2</sup> per year. Similarly, the ClONO<sub>2</sub> changes derived for the same 5-year periods are found equal to 0.08 ± 0.03 and 0.04 ± 0.02 x 10<sup>15</sup> molec./cm<sup>2</sup> per year. Therefore,

the mean [HCl + ClONO<sub>2</sub>] changes over the periods 1984 to 89 and 1991.5 to 96.5 reflect closely the change in the inorganic Cl in the stratosphere, i.e.,  $0.27 \pm 0.05$  and  $0.14 \pm 0.03 \times 10^{15}$  molec./cm<sup>2</sup> per year. This decrease is in line with the evolution of tropospheric chlorine loading [Elkins et al., 1993; Montzka et al., 1996], when accounting for a 4 to 5 year time delay for tropospheric changes of long lived Cl-bearing gases to reflect in the stratosphere. Extrapolation of the 2nd order changes found here suggests that the inorganic Cl loading in the stratosphere will reach a maximum around 2001-2002; this is in good agreement with model predictions based on the most realistic phase-out scenarios of the CFCs and HCFCs as requested by the Montreal Protocol and its amendments. Correlations of the chlorine and fluorine (involving HF and COF<sub>2</sub>) budgets will help to better relate changes as observed here with anthropogenic releases at the ground of CFC substitutes, in particular HFCs and FCs.

### Intercomparison and Validation Activities

Since 1990, the ISSJ has been the "nucleus" for various technique intercomparisons, namely UV-Vis and FTIR spectrometers, infrared laser heterodyne instruments, microwave emission radiometers, even in situ samplers. These were specifically organised as part of the ESMOS/NDSC programs to assess the relative performances of the systems and evaluate complementarities. In addition, sealed cells containing various amounts of HCl gas circulated among the NDSC stations to assess the precision with which the FTIR instruments retrieved the cells' contents. And finally, retrieval algorithm intercomparison exercises that involved 7 codes and 16 groups from all around the world were organized to evaluate the various codes performances and specificities, and to identify possible relative biasing. Current efforts target approaches to precisely determine the ILS of all FTIRs involved in the ESMOS/NDSC network.

Validation activities in support of space-based measurement programs, have included contributions to UARS-CLAES [Rinsland et al., 1996], UARS-HALOE [Russell III et al., 1996a,b] and more recently GOME [Lambert et al., 1997].

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