

Two infrared laser spectrometers for the in situ measurement of stratospheric gas concentration

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Abstract

This paper presents two infrared laser spectrometers built to measure CH₄, N₂O and CO stratospheric concentrations. Both TDL were designed as a subsystem of a composite airborne instruments package, devoted to the chemical and microphysical diagnostics of stratospheric aerosols, developed in the frame of the Airborne Platform for Earth-observation programme (APE). We present the results of the measurements of N₂O and CH₄ concentration in the stratosphere performed in a polar campaign on board of the Geophysica M55 aircraft.

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1. Introduction

N₂O and CH₄ are two anthropically perturbed gases, of particular importance for radiative processes and climate trends, being the most important greenhouse gases, after CO₂ [1], in fact the lower concentration with respect to CO₂ in the atmosphere is balanced by a higher efficacy as greenhouse gas.

The long lifetime, 120–150 and 10–15 years respectively, and small fluxes to the stratosphere cause the atmospheric concentration to be nearly

constant throughout the troposphere and a strong negative vertical gradient above the tropopause, in the stratosphere. For their characteristics N₂O and CH₄ are also studied as atmospheric tracers.

One of the major issues in stratospheric mixing processes is the temporal evolution of fine scale structure in the atmosphere, such as the filamentary structures that break-off from the polar vortex, or localised intrusion of tropospheric air into the stratosphere. The measurement of N₂O and CH₄ is of crucial importance to classify an encountered air mass as typical for the interior of the polar vortex or of the mid-latitude stratosphere.

This paper describes the design, and some field results of two tunable diode lasers spectrometers (TDLs) installed on the Russian high altitude

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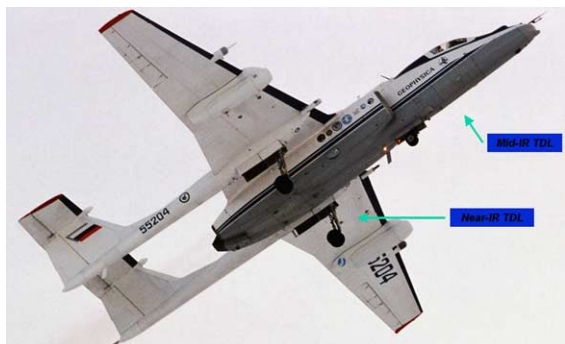


Fig. 1. The Russian high altitude reconnaissance aircraft Myasishchev M55 “Geophysica” and the bay of the two tunable diode lasers spectrometers (TDLs) installed.

reconnaissance aircraft Myasishchev M55 “Geophysica” (Fig. 1).

The M55 Geophysica is currently operated by the European Economic Interest Group (EEIG) Geophysica, and it is now available to the international scientific community. The current scientific payload of the Geophysica is made by 17 different instruments, built by many European research groups, ranging from IR interferometers, UV–VIS DOAS spectrometers, lidars, particle counters, gas chromatographs etc. Even if the same species are normally measured by other instruments, the highly resolved measurements of tracers serve as “yardstick” against which the data of all other instruments can be “clocked” and compared. Without such a tracer it is not possible to determine whether a low ozone value in an air mass sampled by Geophysica is due to the chemical destruction of ozone, or due to the dynamical transport of ozone poor air of low latitude origin to the location of the flight. High precision and spatial resolution data of this tracer will allow detailed investigation of features that are currently under discussion on the atmospheric physicists community.

2. Instruments description

Both TDL spectrometers are based on selective absorption of laser radiation in the infrared (IR) by the molecular species under investigation; one

operates in the mid-IR, around 5 μm , while the other in the near IR between 1.5 and 2 μm . The linear absorption spectroscopy on molecular roto-vibrational spectral lines by TDLs has been widely exploited for the quantitative measurements of trace amounts of several gas species, in many areas: atmospheric chemistry, environmental monitoring, industrial processes monitoring [2–4]. At low pressure as that found at high altitudes around the tropopause (<200 mbar) these absorption bands are resolved in several individual lines, so the technique is quite selective and unaffected by interfering species [3,5].

As all other instruments installed on the Geophysica, they are fully qualified for aeronautic applications, for both mechanical strength and electronic interference [6]. The instruments are also designed to be completely automatic, with an embedded computer providing data acquisition, housekeeping recording and instrument control according the flight profile provided by interaction with avionics through a RS422 interface. The only possible manual operation is an on/off switch on the cockpit that may be operated by the pilot just in an emergency.

Typical flight conditions are between 10 and 22 km of altitude and at an aircraft speed of 160–190 m/s, the operating conditions expected for the TDL are an ambient pressure ranging from 1000 to 30 mbar, a temperature from +70 to –90 °C and strong vibrations.

2.1. Mid-infrared spectrometer

The mid-infrared wavelength region (between 3 and 15 μm) is particularly useful for the detection of the species of interest for atmospheric chemistry, because of the strong absorption bands due to roto-vibrational transitions. At flight conditions, these absorption bands are resolved in several individual lines. The lead salts mid-infrared laser emits at about 2190 cm^{-1} , where both molecules show well resolved roto-vibrational transition lines with no interference from other species and where it emits in single mode with a bandwidth low enough to allow anyway for sub-Doppler resolution spectroscopy of the single roto-vibrational lines. The lead salt laser is mounted in a dewar for liquid

nitrogen cooling and the laser temperature is actively kept constant with an electronic controller. The instrument was then designed for direct absorption measurements with fast sweep integration, inherently self-calibrated for absorption. This technique is usually suitable for absorption levels up to 10^{-3} that would require very long absorption path to achieve the necessary sensitivity.

By adding a current ramp to the injection current the emitted frequency is swept across a frequency interval of about 0.8 cm^{-1} , which is enough to include N_2O and CO simultaneously.

The laser is collimated by a ZnSe lens, then split into a reference and a measurement beam. The first is used for frequency calibration. The second is sent into an astigmatic Herriott-multipass cell, with 182 passes for a total absorption path of 36 m and a volume of 0.31. In normal flight condition the flow is about 4.7 l/min, with a sample clearance of 4 s.

The optical system is enclosed in a thermally insulated and pressurised case in order to have a stable boiling temperature for liquid nitrogen. Fig. 2 shows the box containing the optical system of mid-infrared TDL.

The detection is based on fast scanning of the direct absorption profiles, which are real time averaged at 5 ms/s with a 12-bit resolution. With this technique the concentration can be determined absolutely calibrating the lineshape parameters

with the spectral-lines databases of HITRAN at the measured physical parameters of the sample, i.e. temperature and pressure.

2.2. Near-infrared spectrometer

The near-IR TDL spectrometer, currently measuring CH_4 is named “Airborne Laser Tunable Observer” (ALTO) [7] and was specifically designed to be installed in the unpressurised bay located in the front part of the left wing boom of the M55 Geophysica. The instrument then has to be very compact and lightweight (<30 kg). It is based on a InGaAs/P distributed feed-back (DFB) type diode laser, working at room temperature with thermoelectric temperature stabilisation and emitting at about $1.651 \mu\text{m}$, where the most intense line of a combination roto-vibrational band of CH_4 occur. The distributed feedback (DFB) near infrared lasers exhibit single longitudinal and transverse mode emission, with very narrow bandwidth. The absorption is detected by means of two-tone frequency modulation (TTFM) technique [8] allowing higher absorption sensitivity, as required by the lower line strength in the near-IR. The modulation spectroscopy signal at 2 GHz and the data are processed during the flight.

The laser beam is collimated and split into a reference and a measurement beam: the first is sent through a reference cell filled with 100 mbar of methane for frequency locking of the laser. The position of the reference signal is continuously monitored by the computer and any shift of the laser wavelength is corrected by a control signal to the laser current controller.

The main beam is focused for the measurement in a multipass Herriott cell with base-length of 42 cm, total optical path 31.3 m, and volume 1.09 l.

The instrument is split into two separate cases, one for the optical system (Fig. 3) and one for the electronics, to fit the available space in the aircraft. The gas inlet, built to our specification by MDB, is provided by a 20-mm-diameter probe, located outside the aerodynamic cowl, aimed in the forward flight direction. The time resolution of 1 s corresponds to a spatial resolution of 200 m at aircraft cruise speed.

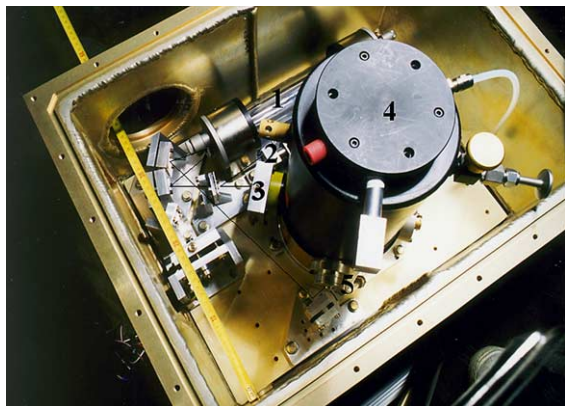


Fig. 2. The thermally insulated and pressurised case with the optical system inside of mid-infrared TDL: (1) multipass cell, (2) reference cell, (3) diode laser, (4) dewar, (5) detector.

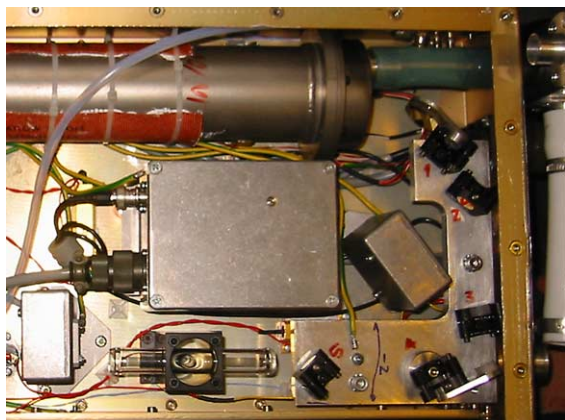


Fig. 3. The case and the optical system inside of near-infrared TDL.

3. Results overview

Both instruments, in the configuration described above, have been operated in the EUPLEX Arctic campaign (Kiruna, 10 flights, January–February 2003, ≈ 60 flight hours), for the study of the polar vortex chemistry and dynamics. As far as the mid-IR instrument is concerned, in these campaigns the CO measurement was of less scientific interest than long time tracers, so we focused our attention on N_2O .

Fig. 4 shows the concentration profile of N_2O measured during the flight on 26 January 2003 in

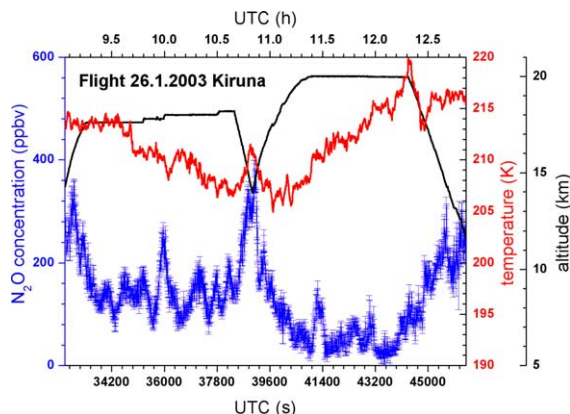


Fig. 4. The concentration profile of N_2O measured during the flight on 26 January 2003 in the EUPLEX campaign in comparison with temperature and pressure profile.

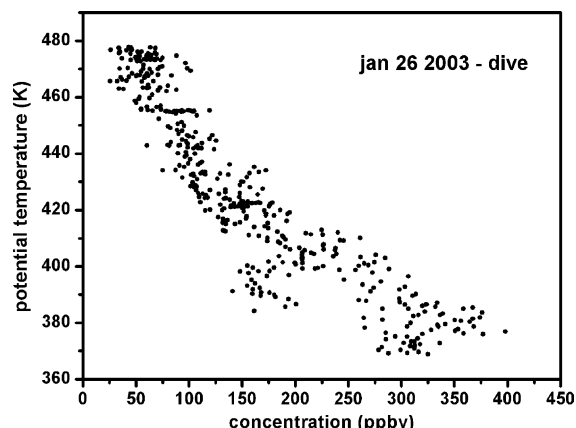


Fig. 5. The concentration profile of N_2O during the plane dive for the same flight in comparison with potential temperature.

the EUPLEX campaign in comparison with temperature and pressure profile. Fig. 5 shows the concentration profile of N_2O during the plane dive for the same flight in comparison with potential temperature.

The main problem in processing N_2O data is the presence of a periodic fringe structure superimposed to the signal. The fringe structure is mainly generated between the reflecting mirrors in the multipass absorption cell, thermally and mechanically induced by the air turbulence. The fringes limited the signal sensitivity for altitude under 10 km.

The scientific significance of the preliminary results is still under investigation.

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References

- [1] IPCC (Intergovernmental Panel on Climate Change), Climate Change (1994), Radiative Forcing of Climate Change and Evaluation of the IPCC IS92 Emission Scenarios, Cambridge University Press, Cambridge, UK, 1995.
- [2] C.R. Webster, R.T. Mentzies, E.D. Hinkley, in: R.M. Measures (Ed.), *Laser Remote Chemical Analysis*, Wiley, New York, 1988.
- [3] D.J. Brassington, in: R.E. Hester (Ed.), *Advances in Spectroscopy*, vol. 24, Wiley, New York, 1995.
- [4] H.I. Schiff, G.I. Mackay, J. Bechara, in: M.W. Sigrist (Ed.), *Air Monitoring by Spectroscopic Techniques*, Wiley, New York, 1994.
- [5] P. Werle, in: P. Hering, P. Lay, S. Stry (Eds.), *Lasers in environmental and life science-modern Analytical methods*, Springer Verlag, Heidelberg, 2004, pp. 223–243.
- [6] G. Toci, P. Mazzinghi, M. Vannini, *Laser Part. Beams* 17 (1) (1999) 59–67.
- [7] F. D’Amato, P. Mazzinghi, F. Castagnoli, *Appl. Phys. B* 75 (2002) 195–202.
- [8] G.R. Janik, C.B. Carlisle, T.F. Gallagher, *J. Opt. Soc. Am. B* 3 (1986) 1070–1074.