

THE MIPAS BALLOON BORNE TRACE CONSTITUENT EXPERIMENT

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ABSTRACT

A novel cryogenic Fourier transform spectrometer (FTS) has been developed for limb emission measurements in the mid IR-region from balloon-borne platforms. The FTS is a rapid scanning interferometer using a modified Michelson arrangement which allows a spectral resolution of 0.04 cm⁻¹ to be achieved. Solid carbon-dioxide cooling of the spectrometer and liquid-helium cooling of the detectors provide adequate sensitivity. The line of sight can be stabilised in terms of azimuth and elevation. A three-mirror off-axis telescope provides good vertical resolution and straylight rejection. Calibration is performed by high elevation and internal blackbody measurements.

Four balloon flights were performed, two of them during spring turn-around 1989 and 1990 over mid-latitudes (Aire sur L'Adour, France, 44°N) and two near the northern polar circle in winter 1992 (Esränge, Sweden, 68°N). Limb emission spectra were collected from 32 km to 39 km floating altitudes covering tangent heights between the lower troposphere and the floating altitude. The trace gases CO₂, H₂O, O₃, CH₄, N₂O, HNO₃, N₂O₅, ClONO₂, CF₂Cl₂, CFCl₃, CHF₂Cl, CCl₄ and C₂H₆ have been identified in the measured spectra. The 1989 data have been analysed to retrieve profiles of O₃, HNO₃, CFCl₃ and CF₂Cl₂. The flights over Kiruna have provided the first ever reported profile measurements of the key reservoir species ClONO₂ and N₂O₅ inside the polar vortex.

1. INTRODUCTION

In recent years the relevance of atmospheric trace gases to the climate system and the ozone chemistry has become known also to the general public. Although the basis of the physical and chemical processes and interactions has been investigated to some degree, there remains still a lot of open questions. An essential precondition for answering these questions is the simultaneous measurement of chemically coupled trace gases in the stratosphere. Fourier transform spectrometers (FTS) proved to be especially appropriate for this task. The most effective way of studying diurnal variations and polar winter conditions is to detect from a high flying carrier at various angles of elevation the radiation emitted by atmospheric constitu-

ents against cold space. In order to achieve the high sensitivity necessary for this kind of measurement, the instrument has to be cooled and the spectral bandwidths have to be restricted.

The cryogenic MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) balloon experiment specially designed for that task has been developed as a joint project between the Institute for Meteorology and Climate Research (IMK) Karlsruhe and the Meteorological Institute Munich (MIM).

2. SYSTEM CONFIGURATION

The technological requirements imposed on any limb-emission sounding device are multiple and stringent. One of the most important requirements is to achieve sufficiently high sensitivity and spectral resolution to detect the weak emissions of atmospheric gases with an adequate signal-to-noise ratio. A second important demand, resulting from the geometry of limb sounding, is a precise and commandable azimuth and elevation pointing device.

The MIPAS balloon experiment consists of seven main sub-systems: gondola and sensor structure, line-of-sight stabilisation system, telescope, interferometer, dewar optics, on-board electronic data-handling system, and the ground-control system. Cooling of the sensor is provided by plates of solid carbon-dioxide which are placed between the insulation and the inner sensor frame and are removed shortly before launch.

The line-of-sight system provided by DLR (Deutsche Forschungsanstalt für Luft- und Raumfahrt) stabilises the gondola in azimuth direction and is used to scan the horizon as well as to look into space or to an internal blackbody for reference measurements. The input data for stabilising the gondola in azimuth direction and the scan mirror in elevation is provided by a set of magnetometers, rate gyros and inclinometers. Additionally, a star-camera looking into the same direction as the line of sight provides an absolute a posteriori reference. Azimuth direction and elevation can be controlled and commanded from ground.

A three-mirror off-axis telescope diminishes the aperture by a factor of approximately 3.7 and reduces the amount of straylight. It provides a geo-

metrical vertical resolution of better than 3 km at the limb.

A double-pendulum interferometer (see Burkert et al., 1983 for a lab version) has been specifically designed for cryogenic environment. One complete interferogram is recorded within 5 seconds providing a spectral resolution of 0.04 cm^{-1} unapodized. The radiation is detected by two liquid He-cooled Si:Ga detectors. The evaluation of laboratory blackbody spectra, with the blackbody at a temperature of 200 K, the instrument cooled and in 1992 flight configuration, showed a noise-equivalent spectral radiance (NESR) of $1.2 \times 10^{-8} \text{ W}/(\text{cm}^2 \text{ sr cm}^{-1})$ in the long-wavelength spectral channel ($750 - 1000 \text{ cm}^{-1}$) and of $7 \times 10^{-9} \text{ W}/(\text{cm}^2 \text{ sr cm}^{-1})$ in the short-wavelength channel ($1170 - 1400 \text{ cm}^{-1}$).

Fast telemetry and telecommand links provide real-time instrument control and allow together with efficient quicklook facilities to decide on changes in the measurement scenario.

A more detailed technical description can be found elsewhere (Friedl-Vallon et al., 1992).

3. BALLOON OPERATIONS AND MEASUREMENT PROGRAMME

Balloon operations were carried out by the CNES (Centre National d'Etudes Spatiales) balloon facility at Aire-sur-l'Adour in the south-west of France and at the Swedish Space Corporation base in Esrange, northern Sweden.

The first flight took place during the night from 17th to 18th May 1989, from Aire-sur-l'Adour. Data were recorded by viewing from 33 km float altitude at tangent heights from the middle troposphere up to nearly floating altitude. During the first flight, the MIPAS instrument was equipped with a single detector whose optical bandwidth was restricted to cover the 690 to 960 cm^{-1} region within the atmospheric infrared window. The unapodized spectral resolution was 0.05 cm^{-1} .

During the second flight in the night from 4th to 5th May 1990, a ceiling altitude of about 39 km was reached. Data were recorded at 20 different elevation angles providing a sampling distance at tangent height of less than 3 km between the upper troposphere and the float altitude. Two detectors with block filters covered the spectral regions 770 to 970 cm^{-1} and 1170 to 1400 cm^{-1} . The unapodized spectral resolution was 0.04 cm^{-1} .

The third and fourth flights were performed as part of the EASOE (European Arctic Stratospheric Ozone Experiment) campaign in January and in March 1992. Both flights were carried out when northern Sweden was inside the vortex boundary. During the flight on January 13 the lower stratosphere ($80 - 40 \text{ hPa}$) was principally cold enough to enable NAT formation. During the flight on March 14 the stratospheric temperatures in the aged vortex were above 200 K.

The flights in the Arctic provided a wealth of spectra for tangent heights between the upper troposphere and about 30 km for different azimuth directions. During or shortly after cut-down operation of the last flight, the shaft of the azimuth decoupling

system broke, and the gondola came down in free fall from 32 km altitude. Upon impact on a frozen lake near Nikel, Russia, the whole instrument was completely destroyed.

4. DATA EVALUATION

The data analysis of limb emission spectra obtained from Fourier transform spectrometers is complex and time consuming. We can only outline here some major steps of the whole chain of data evaluation (for more details see Oelhaf et al., 1991; v. Clarmann et al., 1992).

The housekeeping data are analysed to evaluate the behaviour of the instrument during the flight and to get accurate elevation and azimuth pointing information. The onboard sensors for temperature and pressure as well as radiosoundings from adjacent stations are used to construct a multi-level atmospheric model. The interferograms are quality-controlled and phase-corrected using a short, two-sided interferogram. Deep space and blackbody measurements provide the offset and the gain required to calibrate the raw atmospheric spectra.

The calibrated spectra are analysed by fitting simulated spectra to the measured ones. For this, a retrieval software package inclusive of various optimisation techniques is used, e.g. several implementations of non-linear least-squares fit methods (v. Clarmann, 1990). Normally, the first step is to check the spectra for frequency shifts, spectral resolution, baseline offset and pointing information. Whenever necessary, these parameters are corrected by utilizing the information contained in the spectra as derived from the fitting results. The simulated spectra are calculated by an appropriate level-by-level and line-by-line algorithm (see e.g. Fischer et al., 1988) using most recent spectroscopic data (e.g. Rothman et al., 1987). Trace gas profiles are retrieved from a sequence of atmospheric spectra using the onion peeling method in which the spectra are evaluated successively from the uppermost to the lowest tangent heights.

5. OBSERVED DATA

Examples of MIPAS-B spectra illustrating the quality of the data and a few of the many interesting spectral features observed are presented in Figure 1.

Figure 1a shows an overview of the complete first channel recorded from 39 km altitude viewing a tangent height of about 10 km (May 1990). Depending on its molecular structure, each species has its own characteristic "fingerprint", e.g. the regularly spaced lines of the linear CO_2 molecule, the closely spaced clustered lines of O_3 underlying the CO_2 bands at the left and the right sides of the spectrum, the prominent ν_5 and $2\nu_9$ HNO_3 bands with their two Q-branches, and the broad features of the "heavy" freons, which are composed of many thousands of unresolved lines. In addition, some H_2O lines, marked by dots, have been identified. In this spectral region many species which are relevant to ozone chemistry as well as to the greenhouse effect, are measured simultaneously. Nitric acid and chlo-

rine nitrate, for example, are two key components of the heterogeneous chemistry which are responsible for denitrification and the release of active chlorine which subsequently participate in ozone destruction in the stratosphere of polar regions.

Figures 1b and 1c show selected spectral intervals of Fig. 1a which are subsequently expanded to demonstrate the spectral resolution and sensitivity of the MIPAS-B instrument. The spectrum in Fig. 1c is dominated by a series of Q-branches of the ν_6 fundamental and of some combination bands from three different isotopes of CFC-12 as well as some lines of the CO₂ laser band.

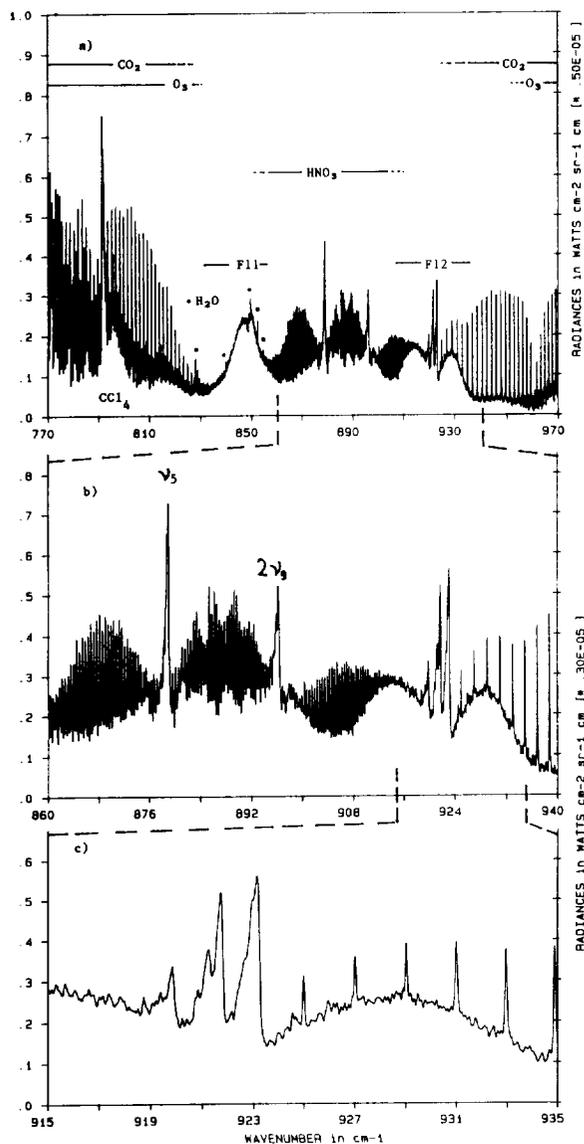


Fig. 1 Limb emission spectrum recorded in channel I during the 1990 balloon flight from 39 km floating altitude. Trace b) and c) show expanded views of subsequently smaller intervals.

The wealth of information contained in the MIPAS balloon spectra can be illustrated by looking at complete limb sequences, which reflect directly the altitude-dependent abundance of various trace gases. Examples of limb sequences measured with MIPAS-B can be found in Oelhaf et al., 1992 and Friedl-Vallon et al., 1992. The complete set of species identified in the long wavelength channel includes CO₂, H₂O, O₃, CCl₄, CFC1₃, CF₂Cl₂, CHF₂Cl, C₂H₆, HNO₃ and ClONO₂.

In channel II (1170 - 1380 cm⁻¹) the species O₃, CO₂, H₂O, CH₄, N₂O, HNO₃ and N₂O₅ have been identified so far. The most interesting data in this region are those connected with N₂O₅. Only very few measurements of this important reservoir gas have been reported. Unequivocal detections of N₂O₅ were made during the flights in 1990 and 1992 (see Oelhaf et al., 1991).

6. PRELIMINARY RESULTS

The data analysis of the four flights is at different stages of progress. The 1989 data have been almost completely evaluated and a few results will be discussed below. Retrieval calculations for trace gas distributions of the 1990 data are in progress. For the EASOE flights preliminary quick-look analysis have been performed for ClONO₂ and HNO₃.

Retrieval results of the 1989 data are shown in Fig. 2 for CFC-12 and O₃ and compared to results of ATMOS (Zander et al., 1987; Gunson et al., 1990) and a set of measurements and model runs for mid-latitudes (Oelhaf et al., 1992). Ozone has been analysed in the 810 cm⁻¹ region by onion peeling, layer-by-layer, according to the corrected tangent altitudes. The CFC-12 profile above 21 km has been scaled as a whole by analysis of its bands at 920 cm⁻¹, because these tangent heights did not contain enough information about CFCs to allow their altitude distributions to be retrieved.

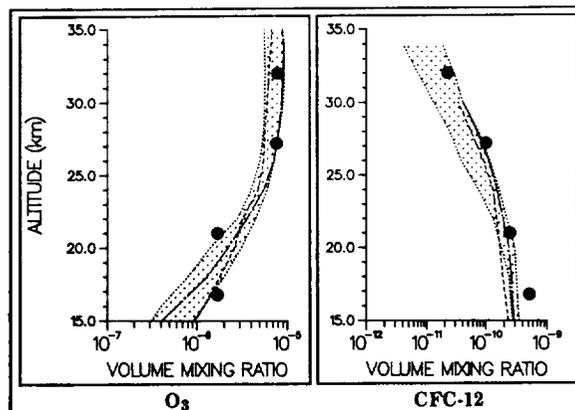


Fig. 2 Retrieved altitude distributions of O₃ and CFC-12 from MIPAS-B 1989 data (●) compared to results from ATMOS and a set of midlatitude summer measurements and photochemical model calculations (see also Fig. 3).

Figure 3 presents the final 1989 data analysis for HNO_3 and a quick-look analysis of the MIPAS data recorded on January 14, 1992 over the Gulf of Bothnia inside the polar vortex. The HNO_3 profile has been retrieved from the P- and R-branches of its ν_5 and $2\nu_9$ bands at 870 and 910 cm^{-1} . Error bars for the 1989 data are derived from extensive Monte Carlo runs for various spectral microwindows taking into account the uncertainty of e.g. altitude of observer position, pointing information, and atmospheric temperature. For the 1990 and 1992 data the error bars are expected to be significantly smaller because of the improved sensitivity of the instrument and the better reliability of the pointing system. The great amount of HNO_3 in the lower stratosphere in the polar winter measurement is qualitatively correlated with a reduced N_2O_5 amount in the same altitude region. This indicates that no significant denitrification took place and N_2O_5 might have been converted into HNO_3 . The complete analysis of the 1989 data is being published elsewhere (v. Clarmann et al., 1992).

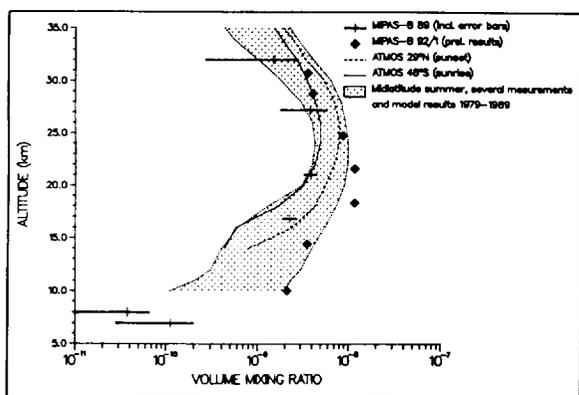


Fig. 3 Retrieved mixing ratios for HNO_3 from MIPAS-B data recorded on May 5, 1989 in mid-latitudes and on January 13, 1992 inside the polar vortex, compared to a set of measurements and model results for midlatitude summer conditions. The analysis for January 1992 is preliminary. (For details, see text).

7. CONCLUSIONS

We have reported on the four flights made within the framework of the MIPAS balloon experiment which yielded a wealth of useful infrared spectra. Trace gases identified so far include CO_2 , O_3 , H_2O , N_2O , CH_4 , CF_2Cl_2 , CF_3Cl , CHF_2Cl , CCl_4 , C_2H_6 , HNO_3 , N_2O_5 and ClONO_2 . The simultaneity of the measurements and the detection of the important reservoir species HNO_3 , ClONO_2 and N_2O_5 , both under mid-latitude summer and in polar winter conditions, is particularly useful.

The first vertical profile measurements of ClONO_2 and N_2O_5 inside the polar vortex together with the main source gases of chlorine and nitrogen will contribute to a better understanding of the polar stratospheric chemistry.

It is intended to build a new instrument as soon as funds will be available. While keeping the

proven basic design, improvements will be made towards a smaller instrument which allows simpler handling. The spectral coverage will be expanded to include also the NO and NO_2 bands. The design of the LOS stabilisation system will need review to meet the requirements.

Acknowledgement: This work has been supported by the German Federal Ministry for Research and Technology (BMFT) and the Commission of the European Community (CEC). (The responsibility for the subject of this publication is with the authors).

REFERENCES

- Burkert, P., Fergg, F and Fischer H., 1983, A Compact High Resolution Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), *IEEE Transactions on Geoscience and Remote Sensing*, GE-21, 3, 345 - 349.
- v. Clarmann, Th., 1990, Die Bestimmung von Mischungsverhältnissen stratosphärischer Spurengase aus Emissionsspektren im infraroten Spektralbereich, *KfK-Report 4698*, Kernforschungszentrum Karlsruhe, FRG.
- v. Clarmann, T. et al., 1992, Retrieval of Trace Gas Profiles from MIPAS-B-89 Limb Emission Spectra, submitted to *Appl. Opt.* (Sept. 92).
- Fischer, H. et al., 1988, Intercomparison of Transmittance and Radiance Algorithms (ITRA), *KfK-Report 4349*, Kernforschungszentrum Karlsruhe, FRG.
- Friedl-Vallon, F. et al., 1992, Limb Emission Spectroscopy with the Balloon-Borne Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), submitted to *Proc. International Symposium on Environmental Sensing*, Berlin, Germany, 22-26 June 1992 (SPIE).
- Gunson, M.R. et al., 1990, Measurements of CH_4 , N_2O , CO , H_2O and O_3 in the Middle Atmosphere by the Atmospheric Trace Molecule Spectroscopy Experiment on Spacelab 3, *J.G.R.*, Vol. 95, No. D9, pp 13867-13882 (1990).
- Oelhaf, H. et al., 1991, Remote Sensing of Trace Gases with a Balloon-Borne Version of the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS), *ESA-Publ. SP-317*, pp 207-213 (November 1991).
- Oelhaf, H. et al., Climatology of Trace Gas Distribution, in: Final Report on MIPAS Science Study - Measurements of Atmospheric Parameters with MIPAS, *ESA Report*, ESTEC/Contract No. 9597/91/NL/SF, to be published (1992).
- Rothman, L.S. et al., 1987, The HITRAN database: 1986 edition, *Appl. Optics*, 26, 4058 - 4097; 1991 HITRAN edition, unpublished results.
- Russel III, J.M. et al., 1988, Measurements of Odd Nitrogen Compounds in the Stratosphere by the ATMOS Experiment on Spacelab 3, *J.G.R.*, Vol. 93, No. D2, pp 1718-1736.
- Zander, R. et al., 1987, Infrared Spectroscopic Measurements of Halogenated Source Gases in the Atmosphere with ATMOS Instrument, *J.G.R.*, Vol. 92, No. D8, pp 9836-9850 (1987).