

# VALIDATION OF MIPAS ON ENVISAT BY IN SITU INSTRUMENTS ON THE M55-GEOPHYSICA

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## ABSTRACT/RESUME

First results of trace gas mixing ratios in the stratosphere derived from limb scans of the MIPAS instrument on the ENVISAT satellite (MIPAS-E) have been compared with coincident measurements made with in situ instruments on the high-altitude aircraft M55-Geophysica between 12 and 20 km altitude during two dedicated campaigns in July and October 2002. Although the number of available limb measurements from MIPAS-E for the comparisons was rather small these comparisons allow a first quantitative statement about the MIPAS-E data quality with the early version of the data processor. At this stage, the ozone data from three limb scans in the stratosphere from 22 July 2002 over Italy (tropopause height:  $\theta = 310$  K) agree within  $(8 \pm 8)$  % with the in situ measurements, which is well within the combined error margins (in situ + MIPAS-E). On the same day the agreement between the  $\text{HNO}_3$  data with a mean deviation of  $(20 \pm 6)$  %, and the  $\text{H}_2\text{O}$  data with  $(15 \pm 12)$  % is also within the combined errors. However, the MIPAS-E  $\text{CH}_4$  data from 22 July 2002 deviate by  $(17 \pm 11)$  %, and the  $\text{N}_2\text{O}$  data by  $(22 \pm 17)$  %, both out of the combined errors of about 15 and 10 %, respectively. Although these first numbers look promising, more extensive comparisons are needed for a sound assessment of the MIPAS-E data quality.

## 1. INTRODUCTION

In order to fully exploit global data sets from satellites, such as the ENVISAT data, for science and/or pollution and global change monitoring these data have to be extensively validated. One possible procedure is the validation by comparison with in situ data sampled by aircraft during dedicated campaigns in different areas of the world, seasons, etc. The advantages of validation by comparison with in situ aircraft data are, for instance, that many species are measured at the same time with high accuracies with independent and well characterized and proven techniques. In addition, aircraft allow a very highly coincident sampling with the satellite and the data have a very high spatial and temporal resolution.

In this paper the first comparisons of the standard products from the MIPAS instrument on the ESA-ENVISAT satellite (MIPAS-E) with in situ data from the M55-Geophysica high altitude aircraft during two dedicated campaigns in July and October 2002 based out of Forli, Italy, will be presented. For this first comparison the volume mixing ratios of  $\text{H}_2\text{O}$ ,  $\text{O}_3$ ,  $\text{HNO}_3$ ,  $\text{CH}_4$ , and  $\text{N}_2\text{O}$  at altitudes between approximately 12 to 20 km sampled with the aircraft have been compared with coincident data from four limb scans of the MIPAS-E instrument.

## 2. M55-GEOPHYSICA INSTRUMENTATION

The in situ chemical instruments on board of the M55-Geophysica aircraft, the corresponding measurement principles, typical measurement accuracies, and the responsible organizations are summarized in Table 1. More information about the instrumentation flown on the M55-Geophysica can be found under <http://ape.iro.ei.cnr.it/>.

Table 1. Chemical in situ instrumentation overview of the M55-Geophysica during the ENVISAT validation campaigns in July and October 2002.

Instrument	Meas. Principle	Species	Accuracy	Organization
FISH	Lyman- $\alpha$	H <sub>2</sub> O (total)	6 %	FZ-Jülich (D)
FLASH	Lyman- $\alpha$	H <sub>2</sub> O (gaseous)	8 %	CAO (Ru)
FOZAN	surface chemiluminescence	O <sub>3</sub>	10 %	CAO (Ru), ISAC-CNR (I)
SIOUX	chemiluminescence	NO, NO <sub>y</sub> (NO <sub>2</sub> , HNO <sub>3</sub> )	5-15 %	DLR-IPA (D)
HAGAR	GC	N <sub>2</sub> O, CH <sub>4</sub> , CFCs, SF <sub>6</sub>	1-7 %	U. Frankfurt (D)
HALOX	chem. conv. res. fluoresc.	ClO, BrO, ClONO <sub>2</sub>	20-40 %	FZ-Jülich (D)
GASCOD	DOAS	O <sub>3</sub> , NO <sub>2</sub> , JNO <sub>2</sub>	10-20 %	ISAC-CNR (I)

Briefly, the total water (i.e. the sum of gas-phase and condensed water) is measured by the Fast In-situ Stratospheric Hygrometer (FISH) of FZ Jülich [1]. The instrument is based on the Lyman- $\alpha$  photofragment fluorescence technique and has been operated on several aircraft and balloons in the past decade. Its measurements are compared to a large number of other atmospheric water vapor measurements using remote-sensing and in-situ techniques [2], amongst others to those of the satellite sensors HALOE and ILAS. The instrument is calibrated after each flight using a calibration bench with a frost point hygrometer as a reference instrument. The overall accuracy of atmospheric measurements, i.e. the accuracy of calibration plus the precision of FISH, is 6%.

The Fluorescent Airborne Stratospheric Hygrometer (FLASH) is used for in situ gas phase water vapor measurements in the upper troposphere and lower stratosphere (0.5-200 ppmv) on board the high altitude airplane M55-Geophysica. It is based on fluorescent balloon hygrometer FLASH-B developed in Central Aerological Observatory (Russia) [3,4]. The instrument uses the fluorescent method which is based on the photodissociation of water molecules by Lyman- $\alpha$  radiation from a hydrogen lamp ( $\lambda = 121.6$  nm) and subsequent fluorescent relaxation of the excited OH\* radicals at wavelength near 310 nm. The fluorescent radiation which is proportional to the water vapor mixing ratio for stratospheric measurements is detected by an optical system with an interference filter and a photomultiplier in photon counting mode. To avoid the influence of ice particles and to provide an intensive air flow through the measuring chamber the hygrometer FLASH contains a special aspiration system. The measuring chamber is automatically opened at the altitude of 8 km during the ascent of airplane and closed at the same altitude during the descent. The instrument's response time is 1 sec, the accuracy is 8%.

FOZAN is a fast chemiluminescent ozone sensor which uses the chemiluminescent reaction between the ozone in the sample air, and the solid-state sensor, of which the main components are laser dyes -Rodamin B, Rodamin 6G, Kumarin 153. The luminescence intensity is registered by a photomultiplier and is proportional to ozone concentration. The instruments work in a concentration range between 10 - 1000 ppbv with an uncertainty of better than 10 % and a sampling frequency of 0.5 Hz.

The NO and NO<sub>y</sub> mixing ratios are measured by the 2-channel NO-O<sub>3</sub> chemiluminescence SIOUX system operated by DLR. The instrument is similar to the one operated on the DLR research aircraft Falcon [5,6,7]. In one channel the NO is measured directly, whereas in the second channel the single NO<sub>y</sub> species are first converted into NO by the help of an Au converter heated at 300°C with CO (0.2 %) as the reduction agent. The nominal detection limits of the two channels at 1 Hz sampling frequency are 5 pptV for NO, and 15 pptV for NO<sub>y</sub>, the accuracies of the measurements are 5 % and 15 % for NO and NO<sub>y</sub>, respectively.

The High Altitude Gas Analyser (HAGAR) is a new instrument for real-time in situ tracer measurements from stratospheric balloons and the M55-Geophysica aircraft that works fully automated under a wide range of environmental conditions from ground up to altitudes up to 30 km. HAGAR consists of a two channel gas chromatograph currently configured to measure N<sub>2</sub>O, CH<sub>4</sub>, Halon-1211, CFC-12, and CFC-11 every 90 s, and a non-dispersive IR-analyser to measure CO<sub>2</sub> with a time resolution of 10s. The precision of the measurements is generally better than 2%, except 3% for H-1211, and 5-7% for CH<sub>4</sub> during July 2002, when the CH<sub>4</sub> channel was flown for the

first time. The instrument is calibrated in flight every 7.5 minutes with 2 standard gases with an absolute accuracy of 2% or better.

The HALOX instrument employs the chemical conversion – resonance fluorescence technique [8] for the in-situ measurement of ClO and BrO. Briefly, the halogen oxides are converted to the respective halogen atoms by addition of NO in a dual duct fast flow system. The atoms are detected downstream by resonance fluorescence in the vacuum UV spectral region. The instrument consists of one measurement duct operating at ambient pressure and one duct employing a pressure reduction by sucking air through a nozzle. For the October campaign the nozzle was equipped with a heater in order to thermally dissociate chlorine nitrate (ClONO<sub>2</sub>) to form ClO which is subsequently detected along with any ambient ClO. By cycling the heater we can differentiate between ClO and the sum of ClO and ClONO<sub>2</sub>. Apart from the instrument onboard the American ER-2 [9] this is only the second instrument available for the in-situ measurement of this species. The ClO and BrO measurements utilize an elaborate technique referencing in-flight data to laboratory calibrations readily carried out before and after each campaign.

The GASCOD/A4π instrument is the airborne version of GASCOD type instruments designed at ISAC/CNR, Italy for UV-Vis Differential Optical Absorption Spectroscopy (DOAS) and actinic measurements application [10,11]. Since 1993 such instruments successfully operate at several stations: Terra Nuova Bay, Antarctica [12], Mt.Cimone, Italy [13], Stara Zagora, Bulgaria [14]. Deploying a DOAS method, the slant column (*sc*) amounts of several trace gases e.g. NO<sub>2</sub>, O<sub>3</sub>, BrO, OClO (under specific conditions) are retrieved. The obtained *sc* are subsequently converted into vertical column (*vc*) amounts and after that, by means of an inversion method, the profiles of these gases can be derived. The 4π index in the instrument's name stands to indicate the ability for measurements of 2π down-welling (direct and diffused) + 2π up-welling diffused solar radiation, and hence to derive the actinic flux.

GASCOD/A4π contains five optical inputs: three deployed for DOAS measurements – one pointed to the zenith, and other two - dedicated for Limb Absorption Measurements (LAM) in perpendicular left and right directions relatively to the flight direction. From these channels ACILA values (Average gas Concentration Inside the Layer near the Aircraft [15]) can be derived. They appear very appropriate quantities for satellite data validation. One of the LAM channels can be switched optically to the nadir (it is foreseen for the next APE/ENVISAT campaign), so trace gas profiles can be splitted into two parts: above and below the aircraft altitude. All three DOAS channels have a narrow field of view (*fov*) of  $1.1 \cdot 10^{-5}$  sr. The collected radiation from the 2π sr *fov* receivers is conducted to the instrumental spectrometric unit by means of optical bundles. More details regarding actinic measurements aboard M55-Geophysica can be found in [16].

### 3. M55-GEOPHYSICA INTERNAL DATA QUALITY CHECKS

In order to verify the quality of the in situ data obtained during the M55-Geophysica campaign in July 2002 the well established correlations between N<sub>2</sub>O and NO<sub>y</sub> as well as O<sub>3</sub> and NO<sub>y</sub> have been calculated and compared with data from former measurement campaigns [17,18]. Panel A of Figure 1 shows the HAGAR N<sub>2</sub>O data plotted against the SIOUX NO<sub>y</sub> data together with the linear fit. Also included is the average correlation ( $\pm 1\sigma$  in red) from [17] derived from ER-2 measurements in the Northern hemisphere, corrected for the N<sub>2</sub>O increase until the year 2002 [19]. It should be noted that this correlation exhibits a maximum change in slope of 7 % between October and January [17]. According to Fig. 1A the M55-Geophysica NO<sub>y</sub> and N<sub>2</sub>O data are well within the variation of the NASA data.

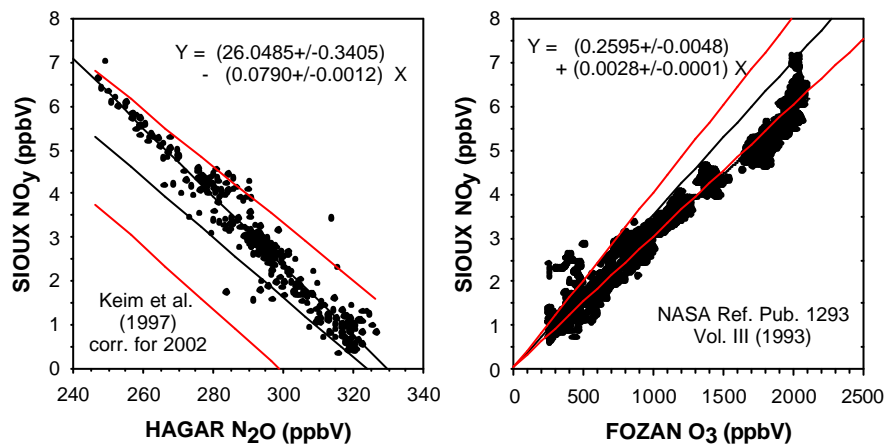


Fig. 1. N<sub>2</sub>O-NO<sub>y</sub> and O<sub>3</sub>-NO<sub>y</sub> correlations from the four M55-Geophysica flight in July 2002 and comparison with data from former campaigns.

Another well established correlation in stratospheric air masses is the slope of the  $\text{NO}_y/\text{O}_3$  ratio which depends on the latitude. The  $\text{O}_3$  vs  $\text{NO}_y$  plot from the SIOUX and FOZAN instruments on the M55-Geophysica for the four flights during the July 2002 campaign is shown on panel B of Fig. 1. Again, the average linear fit of the ER-2 measurements at  $46^\circ\text{N}$  and the standard deviation (in red) is included in the Figure [18], and again the data derived from the measurements on board the M55-Geophysica are within 1 standard deviation of the data found during former US campaigns at comparable latitudes.

A further quality check of the M55-Geophysica  $\text{NO}_y$  data is the comparison of the results with the climatological mean as derived from NASA ER-2 flights during the summer months June, July, and August at  $46^\circ\text{N}$  [20] ([hyperion.gsfc.nasa.gov/pub/atmos/strahan/noy\\_climat](http://hyperion.gsfc.nasa.gov/pub/atmos/strahan/noy_climat)). The data of the four July 2002 validation flights, together with the climatology are shown in Fig. 2 with the potential temperature as the altitude coordinate. Here the M55-Geophysica  $\text{NO}_y$  measurements seem to be slightly lower than the ER-2 climatology but agree within the estimated uncertainties of the  $\text{NO}_y$  measurements of about 12 %.

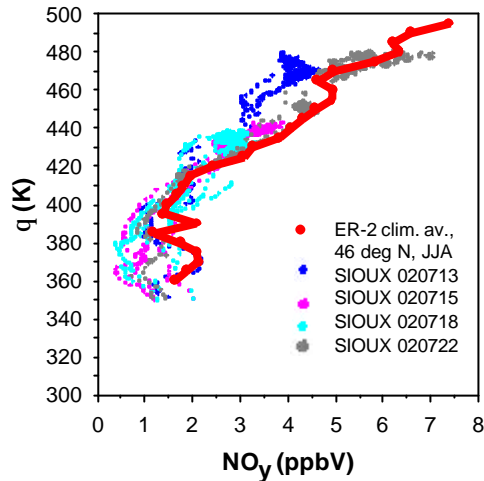


Fig. 2. Altitude profiles of  $\text{NO}_y$  as measured in July 2002 on board the M55-Geophysica compared to a  $\text{NO}_y$  climatology derived from ER-2 flights at Northern midlatitudes in summer [20].

#### 4. COMPARISON OF M55-GEOPHYSICA DATA WITH ENVISAT-MIPAS (MIPAS-E) DATA

Since, at the time of the validation workshop, MIPAS-E data (software version 4.53) for the comparisons with flights from the two M55-Geophysica campaigns were only available for 22 July 2002 and 24 Oct. 2002 ( $\text{H}_2\text{O}$  only) only data from these flights could be used for the first comparisons. The flight track of the M55-Geophysica and the geolocations of the MIPAS-E limb scans (RECORDS) on 22 July 2002 at 0920 UTC over Italy are shown in Fig. 3. The three MIPAS-E limb scans in the vicinity of the aircraft flight track are indicated by their corresponding record number during orbit number 2051. The highest tangent heights of MIPAS-E were measured on the northern points of the records. In the following Figures the data from the different records will be shown in the same color code as in Fig. 3.

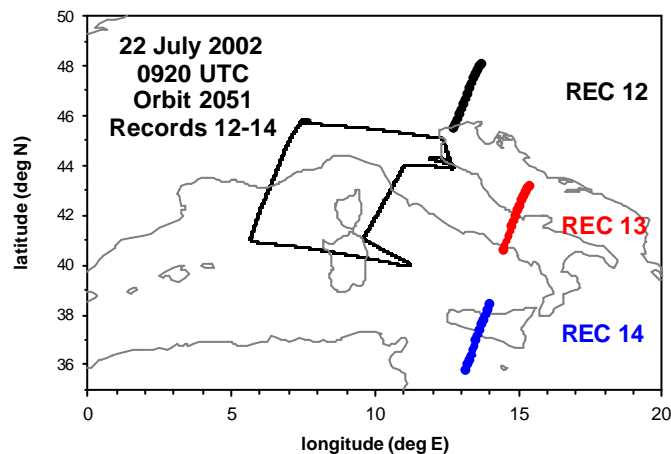


Fig. 3. Aircraft and MIPAS-E measurement locations on 22 July 2002.

#### 4.1 Total and gaseous water (H<sub>2</sub>O)

The water vapor measurements of the FISH instrument on 22 July 2002 during the last part of the flight and the descent of the M55-Geophysica are shown in the left panel of Fig. 4. Also included in this Figure are the corresponding H<sub>2</sub>O mixing ratios as derived from the three scans of MIPAS-E in the stratosphere. The comparison of the MIPAS-E data with the in situ FISH data, excluding the lowermost MIPAS-E datapoint of REC 13 gives a mean deviation of about  $(15 \pm 10)$  % for the satellite data which is still in the range of the combined error bars of the instruments of about 25 %. The scatter in the scans of MIPAS-E at lower altitudes ( $< 380$  K) near the tropopause at 350 K shows the typical spatial variability of stratospheric H<sub>2</sub>O mixing ratios in the lowermost stratosphere.

The right hand side of Fig. 4 shows the good agreement of the FISH and FLASH data in the stratosphere during a dive of the M55-Geophysica from 18 to 12 km altitude over Sicily, Italy, on 24 Oct. 2002. Again the mean deviation of the two MIPAS-E measurements above  $\theta = 380$  K is within the combined errors of the instruments. The lowest MIPAS-E data point on 24 Oct. is obviously caused by a retrieval problem and should be checked.

In summary the mean deviation of the stratospheric H<sub>2</sub>O data during the four MIPAS-E limb-scans, excluding the lowest datapoint on both days, is  $(15 \pm 12)$  % and almost within the combined errors.

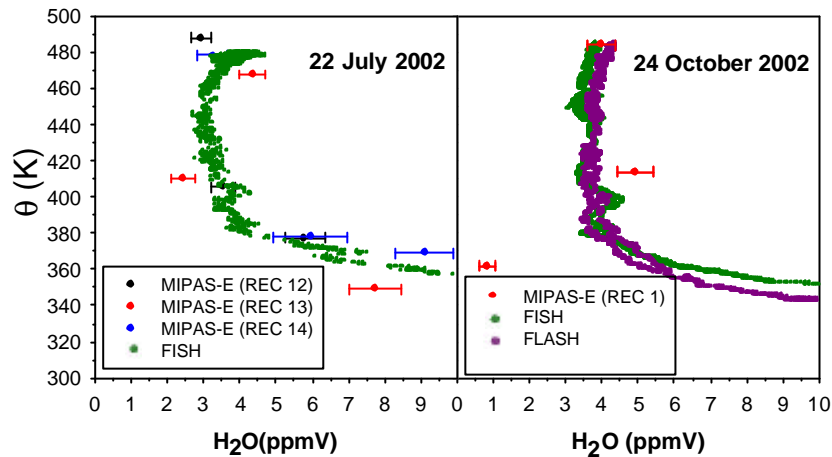


Fig. 4. Comparison of FISH and FLASH on 22 July and 24 Oct. 2002 with MIPAS-E data.

#### 4.2 Ozone (O<sub>3</sub>)

Fig. 5 shows the ozone data from 22 July 2002 from (a) the FOZAN instrument on the M55-Geophysica, (b) the GASCOD instrument on the M55-Geophysica, (c) KNMI TM3-DAM data assimilation model results ([http://www.knmi.nl/gome\\_fd/sciaval/prof/disclaimer.html](http://www.knmi.nl/gome_fd/sciaval/prof/disclaimer.html)) using GOME total column ozone and ECMWF meteorological fields, in the vicinity of Bologna, and (d) the climatological mean over Bologna [21]. The overall agreement of the FOZAN in situ data with the MIPAS-E measurements of ozone is  $(8 \pm 8)$  %. Assuming a combined error for the ozone measurements of about 20 % the deviations are well within these limits.

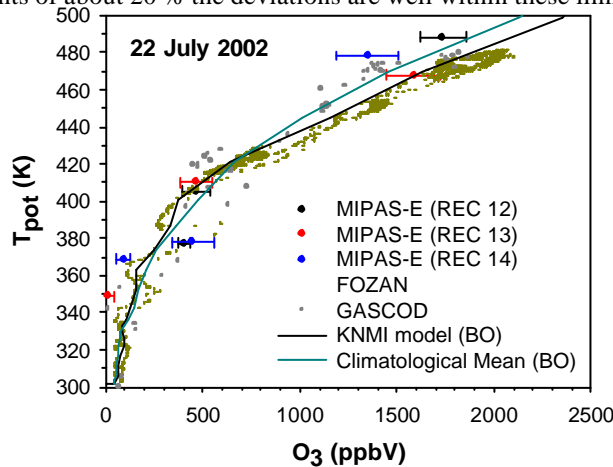


Fig. 5. Comparison of MIPAS-E, FOZAN, GASCOD, model and climatological mean for 22 July 2002.

### 4.3 Nitric Acid (HNO<sub>3</sub>)

The first comparison of HNO<sub>3</sub> mixing ratios derived from MIPAS-E with in situ data from the M55-Geophysica on 22 July 2002 is shown in Fig. 6. Because NO<sub>y</sub> at altitudes between 12 - 20 km consists mainly of HNO<sub>3</sub> ( $\approx 90\%$ ) and, to a minor extent of NO<sub>x</sub> and ClONO<sub>2</sub>, the best way to compare the nitric acid measurements from MIPAS-E with the M55-Geophysica data is to calculate NO<sub>y</sub>-NO<sub>x</sub> from the SIOUX data as shown in Fig. 6. The NO<sub>2</sub> mixing ratios for the NO<sub>x</sub> (=NO+NO<sub>2</sub>) estimation was calculated assuming simple photostationary steady state conditions without accounting for other molecules than O<sub>3</sub> converting NO to NO<sub>2</sub> [22]. The derived mean deviation between the HNO<sub>3</sub> data of the two instruments is (20±6) %, which is within the estimated combined errors of the techniques of about 30 %.

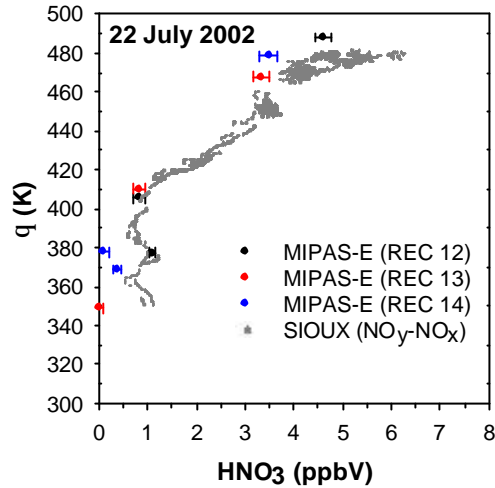


Fig. 6. First comparison of MIPAS-E with SIOUX NO<sub>y</sub>-NO<sub>x</sub> data.

### 4.4 Methane (CH<sub>4</sub>)

The comparison of the in situ methane volume mixing ratios on 22 June 2002 between  $\theta = 300$ -500 K with the three nearest limb scans of MIPAS-E is shown in Fig. 7. The tropospheric mixing ratio below 350 K of 1.773 ppmV for 2002 was taken from [19]. The analysis of the comparison of the stratospheric methane volume mixing ratios reveals a mean deviation of the data of (17±11) % which is out of the range of the estimated combined errors of about 15 % for the CH<sub>4</sub> measurements of the two instruments.

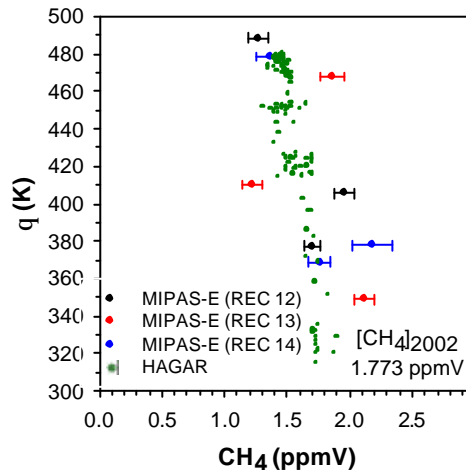


Fig. 7. Comparison of MIPAS-E with CH<sub>4</sub> data from the HAGAR instrument.

#### 4.5 Nitrous oxide (N<sub>2</sub>O)

The N<sub>2</sub>O mixing ratios on 22 July 2002 obtained from the three limb scans of MIPAS-E as well as the HAGAR M55-Geophysica in situ data and a climatological average for summer at 46 deg N taken from [20] and corrected for the year 2002 [19] are shown in Fig. 8. The climatological mean and the HAGAR measurements agree within 5 % at all altitudes. The data from the three limb scans of MIPAS-E do show large variability/oscillations due to retrieval artefacts [23] which leads to deviations from the HAGAR data of (22±17) %. Since the accuracy of the N<sub>2</sub>O measurements on the M55-Geophysica is better than about 7 % this is by far out of the estimated combined error of about 15 %.

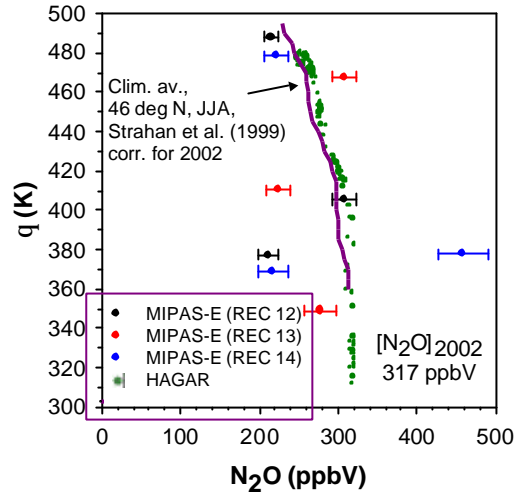


Fig. 8. Comparison of N<sub>2</sub>O data from HAGAR with MIPAS-E on 22 July 2002.

#### 4.6 First measurements of ClONO<sub>2</sub>

The in-situ measurement of ClONO<sub>2</sub> by the newly implemented technique is the only means to independently validate the MIPAS-E measurements of this important species in the lower stratosphere. After extensive laboratory and in-flight tests ClONO<sub>2</sub> could be detected for the first time on the flights on 24 Oct. and 28 Oct. 2002 and the results are shown in Fig. 9. Similar to ClO itself ClONO<sub>2</sub> mixing ratios in mid-latitudes in summer are rather low and only rise steeply around 20 km altitude which is quite well reproduced in the night time measurement of 24 Oct. 2002. The derivation of the daylight profile of 28 Oct. 2002 is not quite as straight forward since during daytime ClO is also present and has to be properly taken care of. For the derivation of these profiles it has been assumed that 100% of the ClONO<sub>2</sub> present in a given air mass is dissociated in the nozzle. For the quantification of the ClO generated by the dissociation the standard ClO calibration is valid. However, the dissociation efficiency of the inlet will be properly characterized in the laboratory within the next few months. For the Arctic ENVISAT validation in March 2003 the measurement of ClONO<sub>2</sub> will be operational.

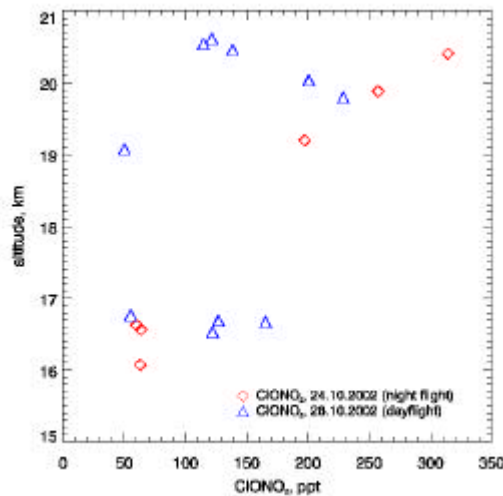


Fig. 9. First in situ ClONO<sub>2</sub> profiles obtained with the HALOX instrument in October 2002.

## 5. SUMMARY AND CONCLUSIONS

Two validation campaigns for the MIPAS instrument on ENVISAT were performed in July and October 2002 with the M55-Geophysica aircraft. Data of the MIPAS-E standard products H<sub>2</sub>O, O<sub>3</sub>, HNO<sub>3</sub>, N<sub>2</sub>O, and CH<sub>4</sub> were successfully collected by the M55-Geophysica in situ instruments during both campaigns.

Internal checks were performed to explore the quality of the in situ measurements on the M55-Geophysica, which included comparisons with climatological averages from previous measurement campaigns at comparable latitudes and seasons in the stratosphere, e.g. from the NASA ER-2 aircraft. Additionally, well known tracer-tracer correlations, such as NO<sub>y</sub>/N<sub>2</sub>O and NO<sub>y</sub>/O<sub>3</sub> were compared and were in agreement with previous observations. From the results of these tests it can be concluded that the M55-Geophysica in situ data are of high quality and are ready for the comparison with available ENVISAT data.

First comparisons of the M55-Geophysica in situ data with the MIPAS-E data were made on the basis of profile data of three limb scans of MIPAS-E on 22 July 2002 and a validation flight on that day in the vicinity of these scans with the M55-Geophysica at altitudes between approximately 7 to 20 km (potential temperature  $\theta = 300$ -500 K). Additionally, H<sub>2</sub>O-data from another limb scan on 24 Oct. 2002 and coincident M55-Geophysica measurements could be compared.

Table 2 gives an overview of the mean deviations ( $\pm 1\sigma$ ) of the ENVISAT-MIPAS data from the in situ results on the basis of the limited number of available coincidences together with the expected single and combined errors of the two instruments. It should be noted that only stratospheric data were considered for the comparison.

Table 2. Overview of the mean deviations and the expected single and combined errors of MIPAS-E and the in situ instrumentation on the M55-Geophysica aircraft. The MIPAS-E uncertainties were taken from [24].

	mean deviation $\pm 1\sigma$ (%)	MIPAS-E uncertainty (%)	expected combined errors (%)
HNO <sub>3</sub>	20 $\pm$ 6	$\approx$ 15	$\approx$ 30 %
O <sub>3</sub>	8 $\pm$ 8	$\approx$ 12	$\approx$ 20 %
H <sub>2</sub> O	15 $\pm$ 12	$\approx$ 20	$\approx$ 25 %
CH <sub>4</sub>	17 $\pm$ 11	$\approx$ 10	$\approx$ 15 %
N <sub>2</sub> O	22 $\pm$ 17	$\approx$ 10	$\approx$ 15 %

For the trace gases HNO<sub>3</sub> and O<sub>3</sub> the comparison shows a good agreement of the data within the expected errors. The deviations between the H<sub>2</sub>O data are a little higher but still within the error margins. However, the comparison of the CH<sub>4</sub> and N<sub>2</sub>O data gives a less favorable picture. The CH<sub>4</sub> mean deviation is almost within the combined errors but the variance reveals a large scatter in the MIPAS-E data. The same is true for the MIPAS-E results for N<sub>2</sub>O, but in addition, the deviation between the two instruments is also larger than the expected combined errors. However, the source of the oscillations in the MIPAS-E altitude profile could be identified by [23] and is due to retrieval artefacts.

Although the number of available limb measurements from MIPAS-E for the comparisons was rather small these comparisons allowed first quantitative statements about the MIPAS-E data quality with the early version of the data processor (V4.53). Although these first numbers look promising, more extensive comparisons are needed for a sound assessment of the MIPAS-E data quality.

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