VALIDATION OF MIPAS N_2O PROFILES BY STRATOSPHERIC BALLOON, AIRCRAFT AND GROUND BASED MEASUREMENTS

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ABSTRACT/RESUME

The ENVISAT validation programme for the atmospheric instruments MIPAS, SCIAMACHY and GOMOS is based on a number of balloon-bone, aircraft and ground-based correlative measurements. In particular the activities of validation scientists were coordinated by ESA within the ENVISAT Stratospheric Aircraft and Balloon Campaign or ESABC. As a companion to a similar paper on CH_4 and in parallel to the contribution of the individual validation teams, the present paper provides a synthesis of comparisons performed between MIPAS N_2O profiles produced by the current ESA operational software (Instrument Processing Facility version 4.61 or IPF v4.61) or by the IMK-FZK scientific processor and correlative measurements obtained from balloon and aircraft experiments as well as from ground-based instruments.

1. INTRODUCTION

As recommended by ESA, validation results presented and discussed during the second Atmospheric Chemistry Validation of ENVISAT (ACVE-2) workshop in May 2004 at ESRIN, Frascati had to be compared with products generated by the latest version of the operational processing software. For the MIPAS N_2O profiles discussed here, the corresponding products were generated by the Instrument Processor Facility or IPF v4.61, but due to the late release and/or incomplete space/time coverage of the corresponding validation dataset, several correlative measurements had to be compared with non-official products. Fortunately, nitrous oxide profiles generated by the IMK-FZK scientific processor have been provided to several validation teams for comparing their own correlative measurements with MIPAS derived profiles. This is especially true for 2003 validation campaigns for which some of the ESA IPF v4.61 products were not yet available by the time of ACVE-2.

The correlative measurements for MIPAS N_2O profiles considered here (see Table 1) have been obtained by balloon experiments (section 2) and by aircraft experiments (section 3) participating in the ENVISAT Stratospheric Aircraft and Balloon Campaign (ESABC) coordinated by P. Wursteisen [1].

Table 1 : ESABC, satellite and ground based contribution to the validation of MIPAS N_2O profiles.

	Instrument	Flight date/campaign period		Latitude coverage	MIPAS dataset available for validation
Balloon	IBEX	28-29 July	2002	Mid latitude	IPF v 4.61
	MIPAS-B	24 Sept.	2002	Mid latitude	IPF v 4.61
	TRIPLE	24 Sept.	2002	Mid latitude	IPF v 4.61
	SPIRALE	2 Oct.	2003	Mid latitude	IPF v 4.61
		21 Jan.	2003	High latitude	IMK-FZK scientific product
	LPMA	4 March	2003	High latitude	IPF v4.61 & IMK-FZK scientific product
		23 March	2003	High latitude	IPF v4.61 & IMK-FZK scientific product
Aircraft	MIPAS-STR	22 July	2002	Mid latitude	IPF v 4.61
	SAFIRE-A	24 Oct.	2002	Mid latitudes	IPF v 4.61
Ground	NDSC FTIR	Fall	2002	High and mid latitudes	IPF v 4.61

Whereas balloon measurements provide high vertical resolution trace species profiles in most of the stratosphere, their specific constraints and limited geographical coverage make aircraft measurements interesting especially for optimising the coincidence or "rendez-vous" possibilities with MIPAS measurements from orbit, but with a smaller vertical coverage of the stratosphere. Since nitrous oxide and methane are passive tracers in the low stratosphere, the availability of simultaneous profiles of these 2 species affords the possibility of internal consistency checks by examining the corresponding CH_4/N_2O correlation plots, which will be discussed in this paper for some of the correlative balloon dataset. Even if a significant effort from the validation scientists and balloon or aircraft operation teams was devoted to reach good space and time coincidence with MIPAS, the number of such correlative data is not yet high enough for a fully significant statistical analysis.

An interesting complementary dataset allowing higher statistics is provided by ground-based profiles of N_2O derived by inversion of atmospheric solar absorption spectra recorded using Fourier transform infrared spectroscopy (FTIR). The vertical resolution of the ground-based data (section 4) is, however, coarser than MIPAS data and averaging kernels have to be used for the comparison. Finally, in section 5, with the caveat that the amount of data available for comparisons is still limited, some preliminary conclusions and recommendations are given.

2. BALLOON-BORNE MEASUREMENTS

The balloon experiments for which N_2O profiles (as well as the corresponding MIPAS data) were available at the time of ACVE-2, include FTIR remote sensing instruments operating in limb thermal emission such as IBEX [2] in the far-infrared and MIPAS-B [3] in the mid-infrared or in solar occultation such as LPMA [4] as well as *in situ* samplers such as the Bonbon cryosampler [5] and *in situ* diode laser spectrometers such as SPIRALE [6]. They are discussed in sequence, a priority being given to the balloon experiments of the 2002 campaigns for which IPF v4.61 MIPAS N_2O profiles are available. In the case of the balloon flights of the 2003 campaigns, some IMK-FZK scientific products (see [7] for a description of the corresponding processor) have been used for comparison when IPF v4.61 data were not available.

2.1 IBEX results

The IBEX (Infrared Balloon Experiment, Istituto di Fisica Applicata "Nello Carrara", IFAC-CNR, Firenze, Italy) [2] is a far-infrared Fourier transform spectrometer, which was flown during the first campaign of ESABC from Sicily (Trapani-Milo; 38 N, 12 E) over the Mediterranean to Spain on 28-29 July 2002. Because there was no coincidence between the period when IBEX was at float and an overpass of ENVISAT, the data used for comparison was taken from MIPAS-E limb scans performed over the Mediterranean within a ± 1 day window covering the IBEX measurements. The corresponding data are plotted in Fig. 1, which shows a reasonable agreement in the mid stratosphere with some dispersion of the balloon data. The MIPAS-E values in the very low stratosphere present a positive bias with respect to IBEX values, a situation which is also seen in other correlative measurements (see below).

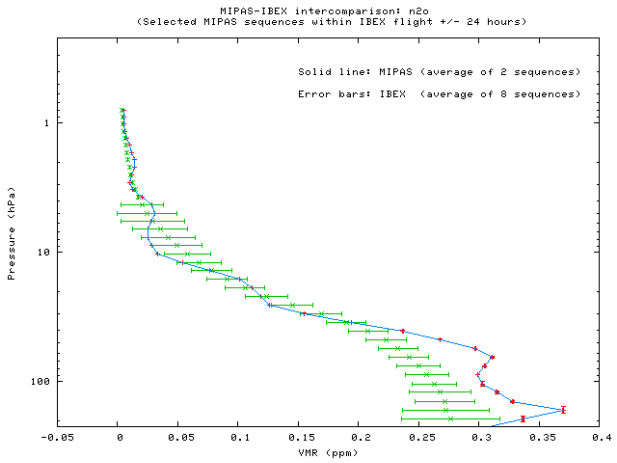

Fig. 1. Comparison of the average of 8 IBEX N_2O profiles (green crosses with error bars) with the average of 2 MIPAS profiles recorded over the Mediterranean within a ± 1 day window covering the float period of the balloon measurements

2.2 MIPAS-B results

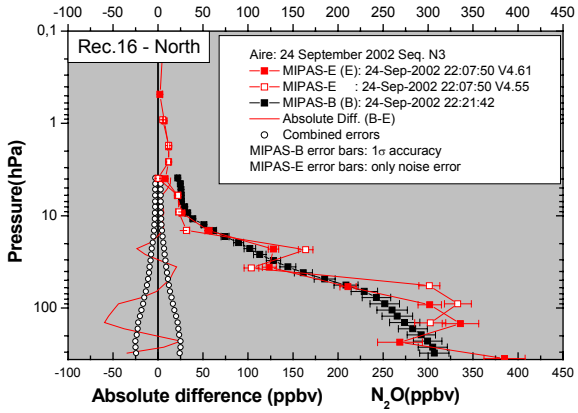

Fig. 2. Validation of MIPAS N_2O v4.55 and v4.61 profiles by MIPAS-B on 24 Sept. 2002 with MIPAS-B minus MIPAS-E v4.61 differences and combined error bars on the left

The MIPAS balloon-borne instrument of Institut für Meteorologie und Klimaforschung, Forschungszentrum Karlsruhe (IMK-FZK), Karlsruhe, Germany, called MIPAS-B [3] is covering exactly the same spectral region as MIPAS-E (the ENVISAT instrument) and is operating in the same observing mode (limb thermal emission). An extremely good space and time coincidence was achieved during the MIPAS-B flight of 24 Sept. 2002 from Aire-

sur-l'Adour (43 N, 0 E). The vertical mixing ratio profiles of N_2O and the corresponding errors are plotted as a function of pressure for the MIPAS versions v4.55 and v4.61 together with the balloon profile. As for CH_4 [8], the "oscillations" observed in v4.55 are significantly reduced in v4.61 (but still present in the lower stratosphere). The differences MIPAS-B minus MIPAS-E v4.61 have to be compared with the combined (root sum squares) error and demonstrate the impact of the remaining "oscillations": the mixing ratio values of MIPAS-E around 300 and 100 hPa are clearly overestimated.

2.3. Cryosampler results

The flight of the cryosampler Bonbon [5] of Institut für Meteorologie und Geophysik, J.W. Goethe Universität, Frankfurt, Germany, took place the same day as the MIPAS-B flight on 24 Sept. 2002, also from Aire-sur-l'Adour.

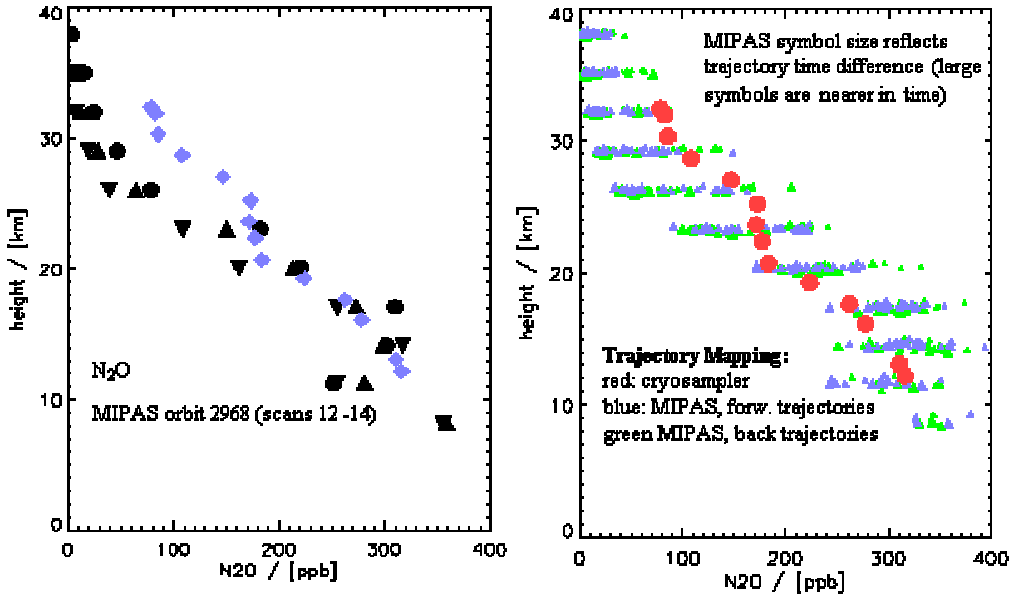

Fig. 3. Validation of MIPAS N_2O v4.61 profiles by the Bonbon cryosampler on 24 Sept. 2002. The left panel is a direct comparison with 3 nearest MIPAS profiles for the same day. The right panel displays 5 days backward and forward trajectory transported profiles for increasing the statistics

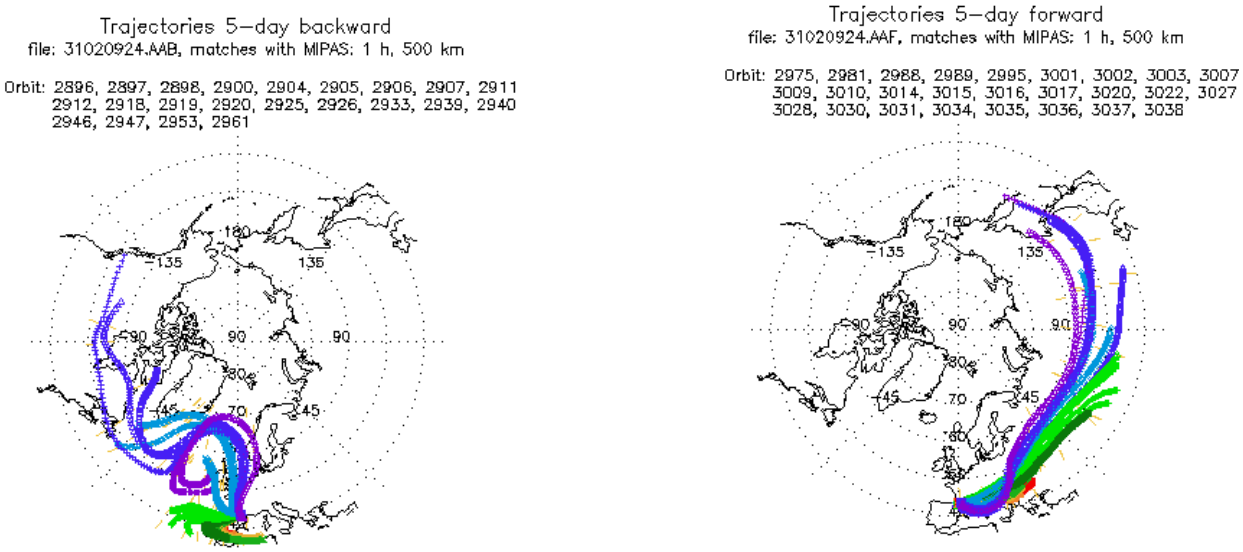

Fig. 4. Trajectory mapping between MIPAS and cryosampler flight data on 24 Sep. 2002. A number of 125 matches (1h, 500 km) is achieved for MIPAS data of 49 orbits within ± 5 days

The v4.61 MIPAS N_2O mixing ratio profiles for 3 limb scans are plotted together with the cryosampler results as a function of altitude on the left panel of Fig. 3. In this comparison and in the following figures care must be taken when MIPAS profiles are plotted as a function of geometric altitude since pointing errors may still affect level 2 IPF v4.61 products. To have a larger statistics five-days forward and backward trajectories shown in Fig. 4 were used to generate the data of the right panel of Fig. 3. In this manner 125 matches involving data acquired by MIPAS over 49 orbits can be "transported" by trajectory mapping to the space and time of the cryosampler measurements. The picture emerging from this comparison is quite satisfactory, with apparently a small negative bias of MIPAS-E in the mid-stratosphere with respect to the cryosampler results. Since simultaneous profiles of the long-lived tracers N_2O and CH_4 (see [8] for the discussion on the validation of the MIPAS profiles for this latter species) are obtained from MIPAS as well as from the cryosampler, it is interesting to check the consistency of the N_2O/CH_4 correlation in both cases. This is shown in Fig. 5 combining the cryosampler results for 24 Sept. 2002 with the MIPAS v4.61 results obtained by trajectory mapping as discussed above. The correlation plot looks reasonable but is confirming the tendency of MIPAS to produce too high N_2O and CH_4 values in the lowest stratosphere.

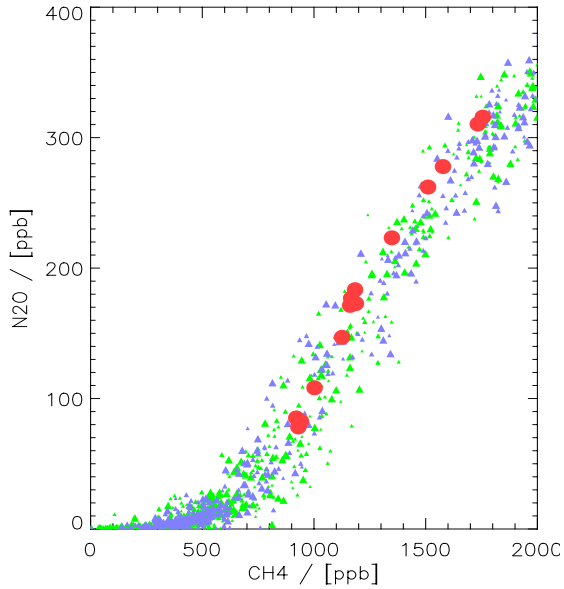

Fig. 5. Comparison of the CH_4/N_2O correlation derived by the Bonbon cryosampler on 24 Sept. 2002 with the MIPAS derived correlation with ± 5 days trajectory mapping

2.4. SPIRALE results

The SPIRALE instrument [6] from Laboratoire de Physique et Chimie de l'Environnement (LPCE, Orléans, France) took place at mid-latitude in the fall 2002 during the ESABC campaign from Aire-sur-l'Adour. Since MIPAS was not operating on 2 Oct. 2002 when SPIRALE was launched, the comparison is only possible with backward trajectories starting from MIPAS measurements on 26, 27 and 28 Sept. and ending at the SPIRALE location on 2 Oct. The N_2O mixing ratio profile of Fig. 6 plotted as a function of potential temperature is demonstrating the capacity of SPIRALE, a fast measurement rate *in situ* diode laser spectrometer, to resolve atmospheric fine structures during ascent (or descent) of the payload. The comparison with the MIPAS values transported by trajectory mapping to the SPIRALE geolocation is reasonable. As for the cryosampler, the simultaneous measurements of N_2O and CH_4 are providing another consistency test when the correlation CH_4/N_2O is plotted (Fig. 7) for the SPIRALE values, for the MIPAS "transported" values and for the reference mid-latitude correlation of ATMOS [9]. Again the high bias of MIPAS for N_2O and CH_4 is observed in the lowest stratosphere.

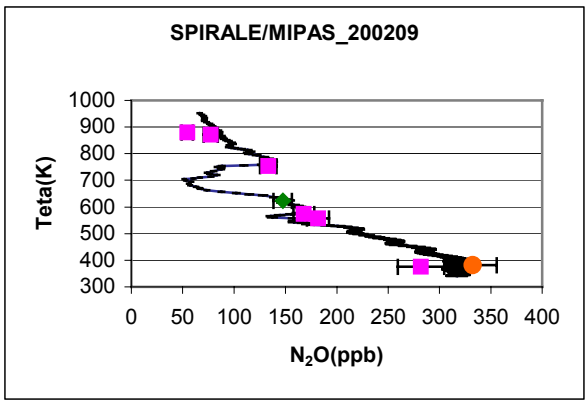

Fig. 6. MIPAS v4.61 and SPIRALE N_2O profiles for the 2 Oct. 2002 ESABC flight from Aire-sur-l'Adour

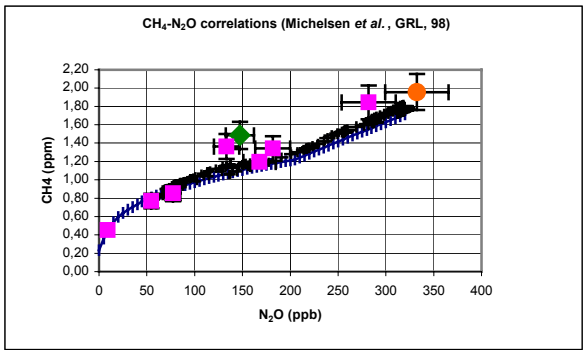

Fig. 7. Comparison of the CH_4/N_2O correlation derived from SPIRALE (black symbols) with the MIPAS correlation (same symbols as in Fig. 6) and with the ATMOS mid-latitude correlation (dark blue)

The SPIRALE instrument was also flown during the winter 2002/2003 ESABC campaign from Esrange (Kiruna, Sweden) on 21 Jan. 2003. The MIPAS v4.61 N_2O profile is not available for that period, but the IMK-FZK scientific processor profile is available and is presented together with the SPIRALE profile as a function of altitude in Fig. 8. The IMK-FZK profile is quite smooth (a difference with IPF profiles due to regularization) and is in good agreement with the SPIRALE profile except in the lower stratosphere where MIPAS N_2O mixing ratios reach unrealistic values (> 350 ppbv).

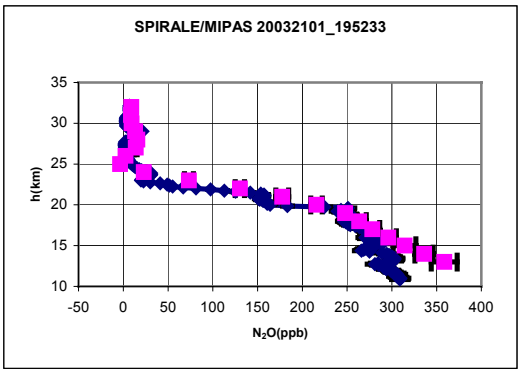

Fig. 8. MIPAS N_2O profile derived with the IMK-FZK scientific processor (purple squares) and SPIRALE data (dark blue diamonds) for the 21 Jan. 2003 flight from Esrange (68 N, 20 E)

2.6. LPMA results

The LPMA balloon-borne FTIR instrument [4] was operating in solar absorption during two flights of the spring 2003 ESABC campaign from Esrange (Kiruna, Sweden). The longwave infrared (LWIR) optical configuration was used on 4 March 2003 (LPMA19) and the shortwave infrared (SWIR) configuration was used on 23 March 2003 (LPMA20). The IPF v4.61 profiles are now available for these dates, but the IMK-FZK scientific products (see [10] for the use of these products in the case of Antarctic ozone hole analyses) were also used for comparing the various N_2O profiles.

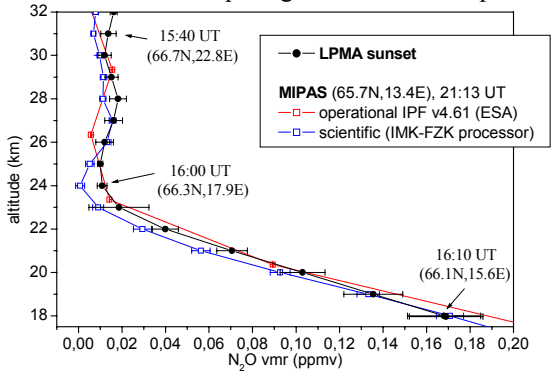

Fig. 9. MIPAS N_2O profiles derived from IPF v4.61 and from the IMK-FZK scientific processor and LPMA balloon measurements obtained on 4 March 2003 during the ESABC campaign from Esrange

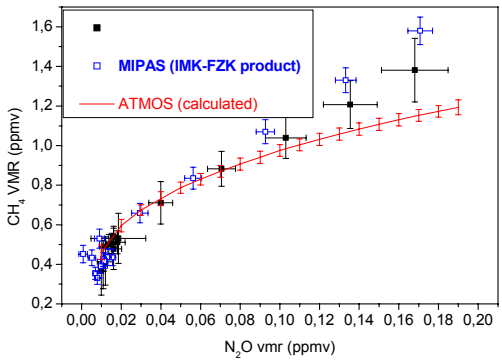

Fig. 10. Comparison of the CH_4/N_2O correlation plots for LPMA (4 March 2003 flight), MIPAS-E (IMK-FZK product) and ATMOS (calculated, outside vortex)

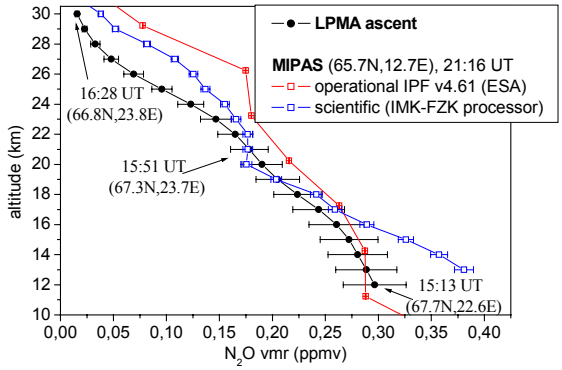

Fig. 11. MIPAS N_2O profiles derived from the IPF v4.61 and from the IMK-FZK scientific processor and LPMA balloon measurements obtained on 23 March 2003 during the ESABC campaign from Esrange

As can be seen in Fig. 9, the agreement between the 3 profiles (LPMA, MIPAS-E from IPF v4.61 and from IMK-FZK scientific processor) is quite good (within the respective error bars) for the measurements of 4 March 2003 in the altitude range 19-31 km.

The correlation plots between CH_4 and N_2O measured by LPMA and MIPAS-E (IMK-FZK scientific product) for 4 March 2003 are compared in Fig. 10 with the calculated correlation [9] derived from ATMOS (outside vortex). The CH_4/N_2O correlation (Fig. 10) is consistent with ATMOS results above 19 km (i.e. for N_2O mixing ratios < 0.1 ppmv), but LPMA and MIPAS values differ from the ATMOS correlation below. The N_2O profiles for the LPMA flight of 23 March 2003 are plotted in Fig. 11 together with the MIPAS-E profiles (IPF v4.61 and IMK-FZK scientific product). The corresponding CH_4/N_2O correlation plots are shown in Fig. 12. A very good agreement is obtained between the LPMA (measured on 23 March 2003) and ATMOS (calculated, outside vortex) curves. The MIPAS results (IPF v4.61 and IMK-FZK products) are clearly departing from the standard correlation curve, however. The structure of the MIPAS-E profile around 26 km (0.17 ppmv for IPF v4.61) does not seem realistic. The shark fin-like structure in the IMK N_2O-CH_4 correlation

(Fig. 12) is caused by different altitude resolutions of these profiles in this particular case. Different response to small-scale structures shows up as an unphysical notch in the correlation curve.

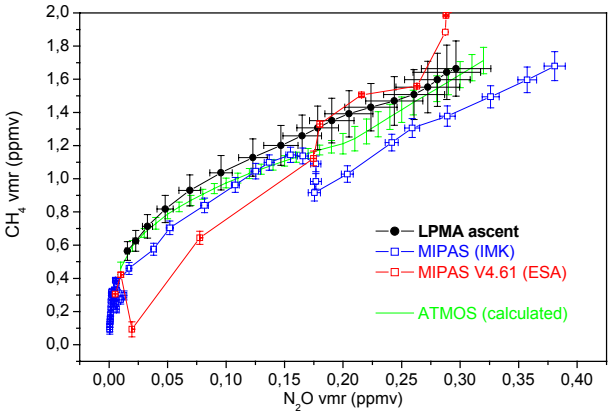

Fig. 12. Comparison of the CH_4/N_2O correlation plots for LPMA (23 March 2003 flight), MIPAS-E (IPF v4.61 and IMK-FZK products) and ATMOS (calculated, outside vortex)

3. AIRCRAFT OBSERVATIONS

Two aircraft instruments providing remote sensing measurements of N_2O have been used within ESABC during different phases of the deployment of the M-55 Geophysica. The first instrument is MIPAS-STR operating in the mid-infrared, the second one is SAFIRE-A operating in the far-infrared. Both instruments are FTIR spectrometers scanning the limb in the thermal emission geometry. As compared to balloon platforms accommodating similar instruments, the flight pattern of the M-55 aircraft can be optimised for good space and time geolocation with MIPAS-E measurements, but the vertical coverage of the aircraft FTIR instruments is reduced (~ 6 to 21 km).

3.1. MIPAS-STR results

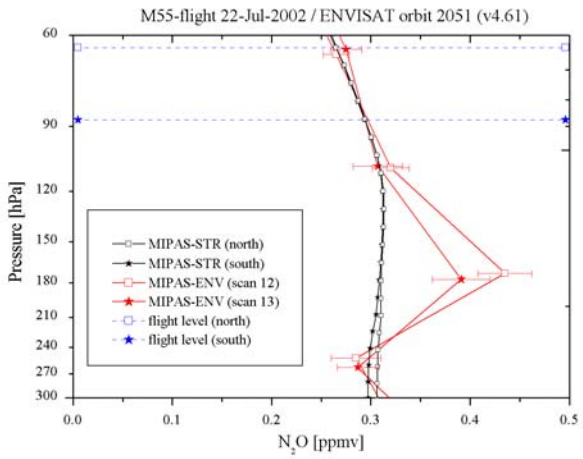

Fig. 13. MIPAS-E N_2O profiles produced by IPR v4.61 and MIPAS-STR measurements acquired on 22 July 2002 from the M-55

The FTIR Instrument MIPAS-STR [11] is operated by Forschungszentrum Karlsruhe, IMK-FZK on the M-55 Geophysica aircraft. Thermal emission limb spectra are recorded from cruise altitude in the same spectral domain as MIPAS-E onboard ENVISAT. The N_2O profiles of MIPAS-STR (two viewing directions, see [11] for more details) are plotted in Fig. 13, together with the coinciding profiles of MIPAS-E. Clearly the high mixing ratio of N_2O for MIPAS-E in the lower stratosphere at ~ 180 hPa is unrealistic and is possibly resulting from the lack of regularization in the IPF v4.61 retrieval algorithm.

3.2. SAFIRE-A result

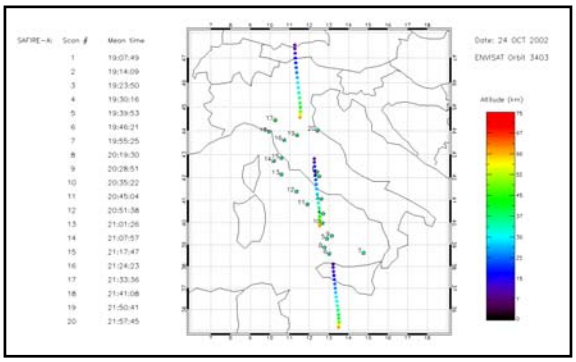

Fig. 14. M-55 Geophysica mid-latitude flight of 24 October 2002: MIPAS-ENVISAT N_2O validation with SAFIRE-A

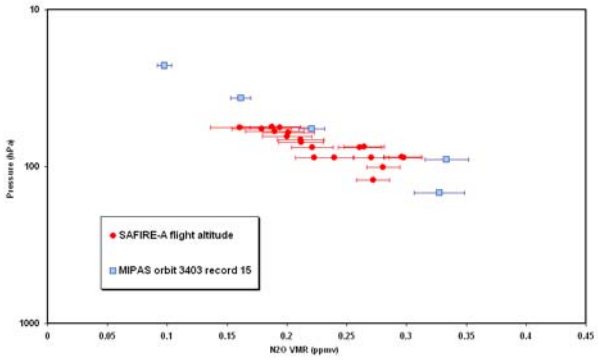

Fig. 15. Comparison of ENVISAT orbit 3403, MIPAS scan 15 N_2O vmr measurements with SAFIRE-A for 24 October 2002

The other FTIR instrument measuring N_2O profiles from aircraft in the upper troposphere/lower stratosphere (UT/LS) is SAFIRE-A [12, 13] (Spectroscopy of the Atmosphere by using Far-Infrared Emission) of Institute of Applied Physics, "Nello Carrara", IFAC-CNR, Firenze, Italy. The geolocation of the SAFIRE-A limb scans and of the corresponding MIPAS-E tangent points is presented in Fig. 14 for the M-55 flight of 24 Oct. 2002, demonstrating the high degree of coincidence between aircraft and satellite measurements. The N_2O mixing ratio values are plotted in Fig. 15 for MIPAS-E limb scan 15 and for the corresponding

SAFIRE-A data. Clearly around the 100 hPa level, MIPAS-E presents a positive bias with respect to correlative measurements as already noticed for other comparisons in the UT/LS.

4. GROUND-BASED RESULTS

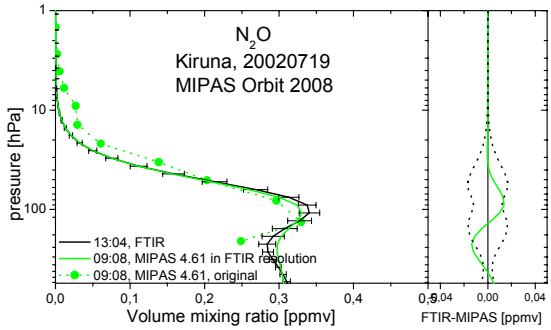

Fig. 16. Comparison of N_2O profiles derived from FTIR ground-based spectra recorded at Kiruna with MIPAS v4.61 profiles (raw = green dots; smoothed = green line)

The validation of MIPAS N_2O profiles with ground-based measurements is difficult but feasible using atmospheric absorption spectra recorded at high spectral resolution (~ 0.002 cm^{-1}) by the Bruker FTIR instruments of the Network for Detection of Stratospheric Change (NDSC). The inversion of the corresponding spectra, which can be recorded for each ENVISAT overpass of the station with the sun as a source in clear sky conditions, is producing mixing ratio profiles with a vertical resolution of about 8 km in the stratosphere [14]. For a rigorous comparison of the ground-based profiles with MIPAS, the smoothing due to the lower vertical resolution of the NDSC FTIR instruments is accounted for using the ground-based averaging kernels (matrix **A**):

$$\mathbf{x}^{smoothed} = \mathbf{x}_a + \mathbf{A}\,(\mathbf{x}^{raw} - \mathbf{x}_a)$$

In this equation $\mathbf{x}_a$ represents the *a priori* profile used in the retrievals, whereas $\mathbf{x}^{raw}$ and $\mathbf{x}^{smoothed}$ represent respectively the MIPAS profile and the corresponding

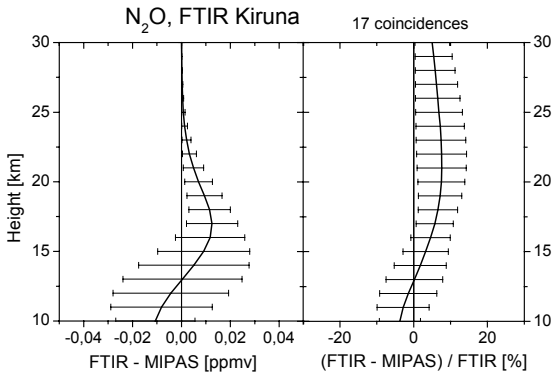

Fig. 17. Mean difference and relative difference between 17 ground-based FTIR N_2O profiles and the corresponding MIPAS profiles

profile suitable for comparison with the ground-based FTIR measurements. An example of single N_2O profile comparison is shown in Fig. 16 for measurements acquired from Kiruna and processed at IMK [14]. A comparison with better statistical significance is presented in Fig. 17 for a set of 17 FTIR profiles in coincidence with MIPAS v4.61 N_2O profiles during the period 18 July 2002 to 1 November 2002. A similar type of processing has been done for FTIR spectra recorded at Jungfraujoch [15] and the results are presented in Fig. 18 for 2 cases in the fall 2002. The raw and smoothed MIPAS profiles appear respectively in the left and right panel together with the FTIR profiles and the *a priori* profiles used in the analyses. A consistency check is also provided when vertical columns are compared (see [15] for a more extensive discussion). This is shown in Fig. 19 where the "stratosphere" columns derived from FTIR spectra in the fall 2002 (the horizontal scale is in modified Julian day MJD 2000) are plotted together with the corresponding MIPAS derived column. The positive bias of MIPAS in the UT/LS is found again here in the higher values of the MIPAS columns as compared to FTIR. But one has to be careful with the exact lower limit for integrating the stratospheric columns.

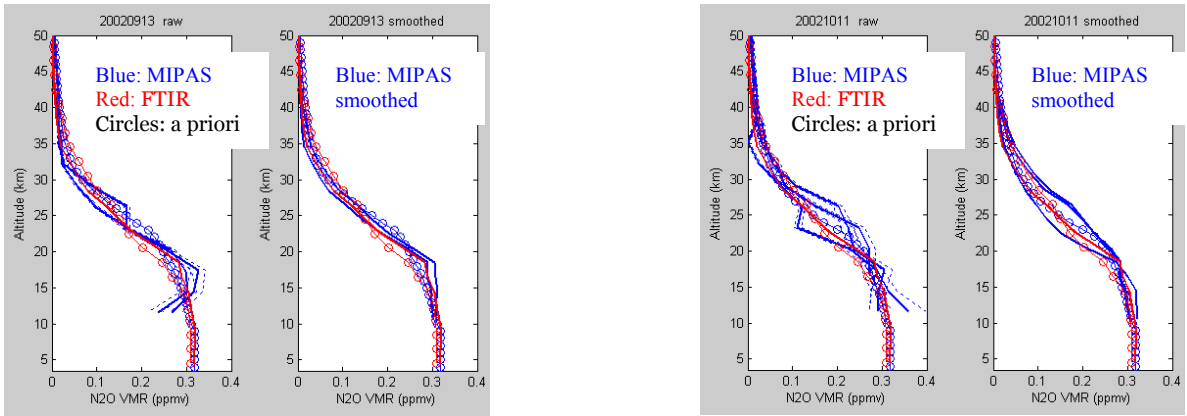

Fig. 18. Comparison of N_2O profiles derived from FTIR ground-based spectra recorded at Jungfraujoch (ISSJ, Switzerland) with MIPAS IPF v4.61 profiles (circle = *a priori* profiles; continuous lines = FTIR in red, MIPAS in blue)

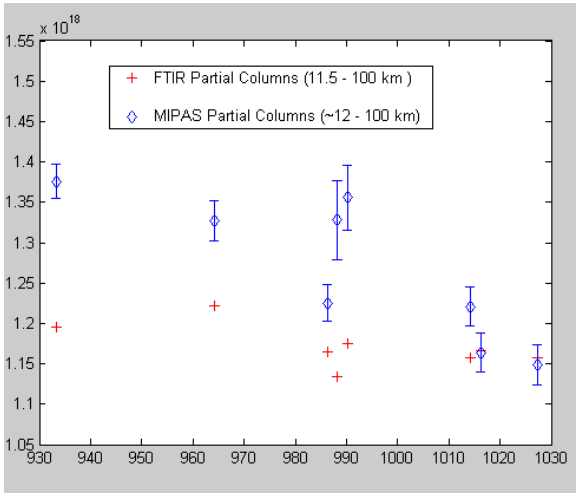

Fig. 19. N_2O vertical columns from ground-based FTIR (ISSJ-Jungfraujoch) compared to the corresponding MIPAS derived columns. Integration over the profiles is performed within the given altitude limits. The horizontal scale is MJD2000

5. CONCLUSION

The MIPAS N_2O profile validation exercise presented here is resulting from comparison of IPF v4.61 or IMK-FZK datasets available at the time of the ACVE-2 meeting with correlative profiles obtained from balloon-borne, aircraft and ground-based instruments. Even if the data for comparison are already significant in number, a firm conclusion is still awaiting further analyses. However, the following findings apply:

- overall MIPAS is indeed measuring N_2O reliably at the 10 % level of precision
- a positive bias of MIPAS with respect to several types of correlative measurements is present in the lower stratosphere/upper troposphere (i.e. for pressure greater than 100 hPa) for N_2O (and CH_4, see [8])
- the comparisons in the lower part of the MIPAS N_2O profiles is often hindered by the weakly constrained IPF v4.61 algorithm or by oscillations generated in the overall MIPAS system
- the CH_4/N_2O correlation is useful to detect spurious MIPAS values in one or the other profile of these long-lived tracers. This check should be done systematically by ESA to flag (and examine later) profiles where the observed correlation is far from expectation
- ground-based FTIR profiles of N_2O, when compared with the appropriately smoothed MIPAS profiles, have demonstrated their potential for higher statistics and mid-term trends in the MIPAS/correlative data comparison.

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