# PROGRESS IN

# MIPAS OBSERVATIONS OF CFC TRENDS

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## ABSTRACT

MIPAS is a Fourier transform interferometer on Envisat, observing the infra-red thermal self emission of the atmosphere.

We investigate the trend in the observed concentrations of CFC-11 ($CCl_3F$) and CFC-12 ($CCl_2F_2$) throughout the life of the instrument. From September 2002 to March 2004, the nominal 'high resolution' (0.025cm$^{-1}$) reprocessed L1B data is used. New work was performed to enable the use of the 'reduced resolution' (0.0625cm$^{-1}$) data now being taken so the trend calculation could be extended to early 2006.

CFCs are implicated in the catalytic destruction of ozone and the polar 'ozone holes'. They are controlled by the Montreal protocol, and this work provides some means of quantifying its effect.

The long time series from a single satellite instrument is useful for tracking the evolution of the CFC stratospheric loading, and checking the accuracy of simulations.

The profiles of these species were determined using 'MORSE', developed at Oxford. It uses optimal estimation. Resulting profiles were combined a posteriori and three-day means calculated on a monthly basis. These means were used to determine the trends in various zonal latitude bands and globally.

## 1. INTRODUCTION

MIPAS was launched as part of Europe's environmental monitoring satellite, Envisat, aboard an Ariane-5 rocket on 1st March 2002. Envisat was injected into a polar orbit at an altitude of about 800 km and has an orbital period of about 100 minutes. MIPAS is an actively cooled atmospheric thermal emission limb sounder, working in the mid-infrared with a field-of-view that is approximately 3 x 30 x 300 km.

MIPAS obtains high resolution spectra (0.025 cm$^{-1}$) that cover the range 685–2410 cm$^{-1}$ (14.6–4.15 µm). A complete limb scan sequence consists of 17 spectra with tangent points at 68 km, 60 km, 52 km, 47 km, 42 km and downward to 6 km in 3 km steps.

The CFCs (chlorofluorocarbons) under investigation are known to have significant anthropogenic sources, and were used extensively as inert refrigeration and blowing agents.

The sink (destruction) processes mainly involve photolysis by short wavelength UV in the stratosphere and mesosphere. They are also involved in radical chemistry.

They have relatively long lifetimes. F11 has a lifetime estimated at 45 years. F12 is longer lived with a lifetime of around 100 years.

The gases are well mixed in the troposphere as a result of the long lifetimes and variability in their tropospheric distribution would be mainly due to the non-uniform distribution of their surface source terms. The volume mixing ratios (VMRs) of the gases are ~250pptv (F11) and ~500pptv (F12) respectively. There is significant stratospheric height and latitude dependence, in line with stratospheric circulation and destruction locations.

In light of the discovery of the ozone hole in the 1980s, there are now treaties governing production and use of these species. Both F11 and F12 regarded as *Greenhouse Gases* (Intergovernmental Panel on Climate Change) and *Ozone Depleting Substances* (Montreal Protocol). As a result, there has been a significant reduction in emission, especially industrialized nations. This paper investigates the results of this major reduction.

Using data from [1], F11 trends over the past 5 years are estimated at -5 pptv yr$^{-1}$ (about 2%). There is no estimated F12 trend, having only stopped increasing in the past couple of years.

## 2. REDUCED RESOLUTION L1B DATA

A change was made to the nominal resolution at which the MIPAS instrument operates. From the start of 2005 all routine measurements have been made at 0.0625 cm$^{-}$

[1]. The difference between the two resolutions is shown for a small region of the spectrum with high sensitivity to F11, Fig. 1. The change was implemented because hardware monitoring observed large numbers of drive unit errors. The use of ‘rest’ periods allows ‘self-healing’ and reduction in error rate. In addition, a reduction in corner cube slide distance was found to reduce the number of slide errors. The rest periods introduce regular, planned breaks in observations and the reduced slide movement reduces interferometer resolution. As a shorter slide path requires less time, measurement patterns were also changed. It is possible to either take 27 scans in the same time as the nominal 17 took (the new Nominal mode), or to sample more profiles (over 100 for the UTLS-1 mode, instead of the 70 normally taken).

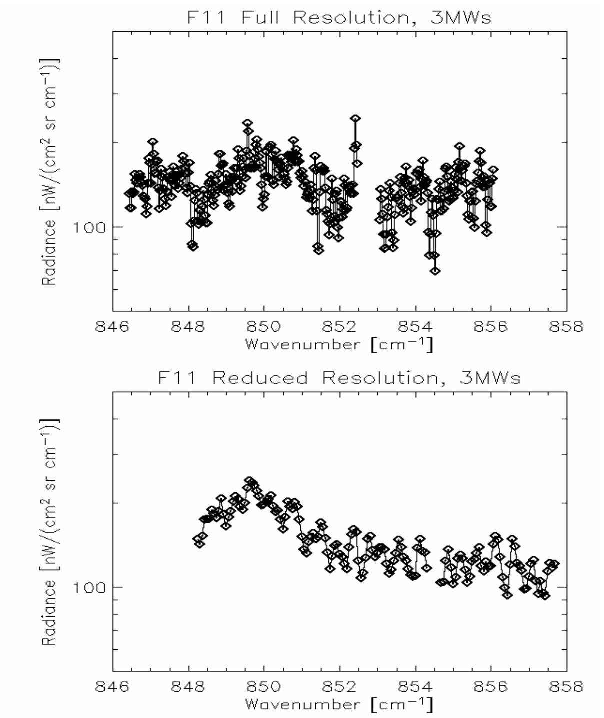


Figure 1: Full and reduced resolution from a single measurement. Note how much smoother the lower resolution spectra appears as atmospheric lines are less well resolved.

## 3. MICROWINDOW SELECTION

Microwindows are small regions of the spectrum no more than three wavenumbers wide. These regions are optimised to maximise the information obtained on the target species, whilst minimising both random and systematic errors. An informative discussion of the development of the microwindow selection algorithm may be found in [2]. Together with the microwindow regions, a detailed error analysis is also obtained, Fig. 2, showing the relative significance of all the known error sources. The retrieval is sensitive to temperature accuracy, pressure accuracy, strong radiance gradients and radiometric calibration.

The reduction in resolution (RR modes) increased the significance of random noise, mainly due to the over sampling strategy (1.5 km step with a 3km FOV). This can be mitigated to some extent by co-addition.

Microwindows have been re-selected for all operational species using reduced resolution parameters.

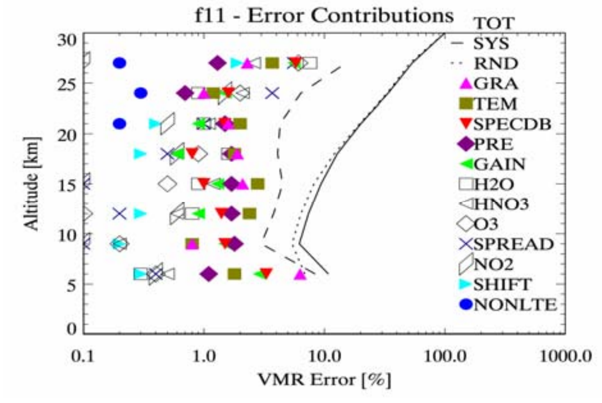


Figure 2: Assignment of various error sources to the final error budget for the full resolution case as calculated during microwindow selection. The total error is shown (solid line) together with the random error component (dotted line) based on the instrument NESR. The total of the systematic error contributions is also shown (dashed line).Significant systematic error sources include: ‘TEM’ – retrieved temperature uncertainty, ‘GRA’ – error from a horizontal radiance gradient, ‘H2O’ – water vapour uncertainty, ‘GAIN’ – radiometric calibration and ‘SPECDB’ – uncertainty in spectroscopic line parameters. The a priori error is assumed to be 100% and its covariance diagonal.

## 4. INTERFERING SPECIES RETRIEVAL

One of the aims of this work was the demonstration of ‘operational’ retrievals using the new nominal scan pattern and resolution. These patterns are listed in Table 1. Some example retrievals of temperature and ozone from a single orbit using the new reduced resolution microwindows are shown in Fig. 3.

Table 1: Comparison of scan patterns.

| **Mode** | **Levels** | **Altitudes [km]** |
|---|---|---|
| **Nominal** | **17** | **68,60,52,47,42,39,36,33,30,27,24,21,18,15,12,9,6** |
| **Nominal RR** | **27** | **70,66,62,58,54,50,46,43,40,37,34,31,29,27,25,23,21,19.5,18,16.5,15,13.5,12,10.5,9,7.5,6** |
| **UTLS1 RR** | **19** | **51,47.5,43,38.5,34,31,28,26,24,21.5,20,18.5,17,15.5,14,12.5,11,9.5,8** |

The full resolution retrieval (using 'MORSE' [3]) is already well validated. The reduced resolution retrieval of the ESA operational species has not been formally validated, especially through collocated measurements from other instruments or the surface. However, quantitative comparison with climatologies and known general structure of the zonal fields of these species suggest the retrieval is working well. We have also compared the error characteristics (averaging kernels, estimated random error, degree of oscillation, a priori contribution to the final profile) with the previously validated full resolution retrievals, with encouraging results.

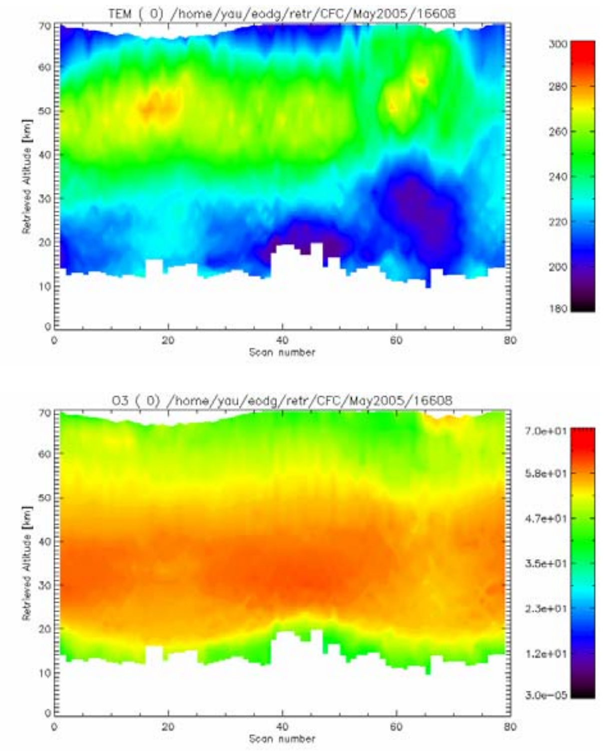


Figure 3: Interfering species retrieval. Zonal mean temperature and ozone from 3 days of measurements in May 2005 retrieved from the new nominal resolution mode (0.0625 cm-1, 27 sweeps per scan). The 'checked' effect originates from a difference in the radiometric calibration of the forward and reverse sweeps.

## 5. RETRIEVED F11 AND F12 ZONAL MEANS

MIPAS measurements (L1B) exist from mid-2002 to Feb 2006. We selected a wide distribution for maximum sensitivity and had to retrieve operational species first.

Blocks of three days in the month were selected as a processing time trade-off. This is sufficient (about 250 scans) to enable good reduction in the random component through co-addition. Figs. 4-6 show the resulting retrieved zonal mean fields for the three measurement types discussed.

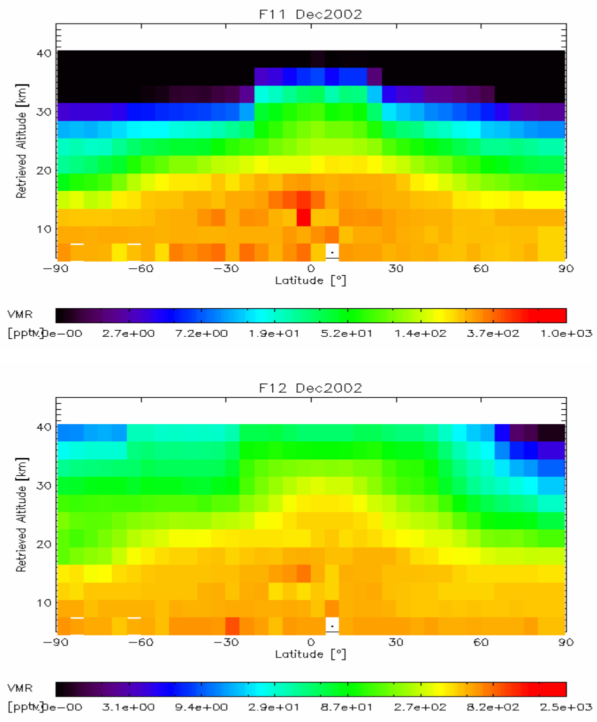


Figure 4: Full resolution F11 (top) and F12 zonal mean fields from December 2002. Descent at the winter pole (low values) is visible. The tropical troposphere contains some residual cloud effects that give rise o some of the isolated high values.

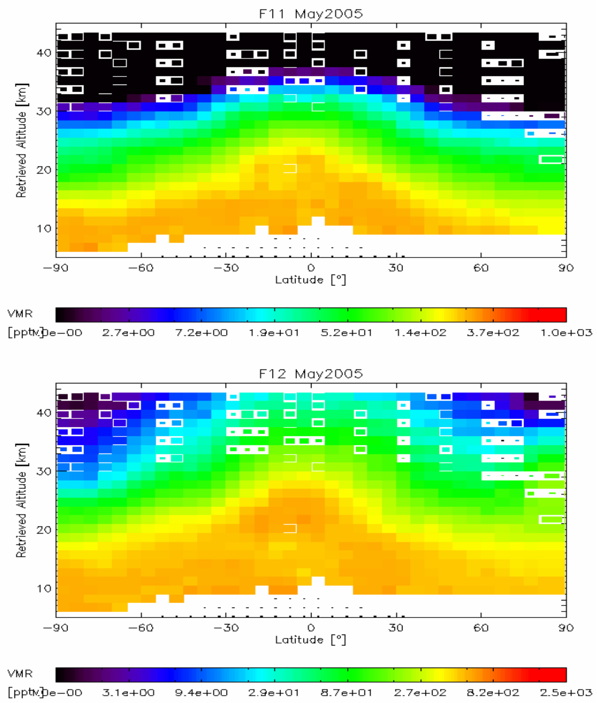


Figure 5: Reduced resolution, new nominal sampling mode for F11 (top) and F12 from May 2005. Asymmetry in the tropical maximum is clearly visible

as is the increased vertical resolution (as good as 2km in the lower areas of the figure).

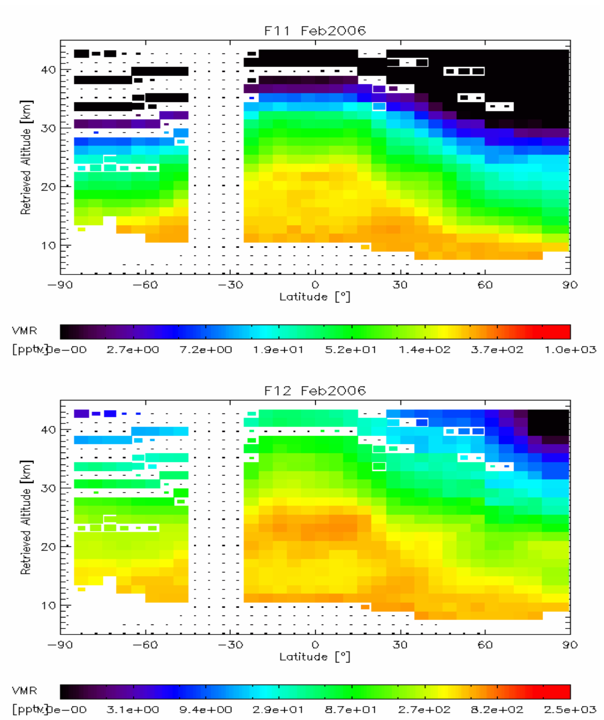


Figure 6: Reduced Resolution UTLS1 mode retrievals of F11 (top) and F12. The data coverage is not so good, but the general distribution of the two species is clearly visible.

## 6. CALCULATING A TREND

The trends were calculated from a simple lower stratospheric (15 - 30km) mean, combining 3 days of measurements each. The random error on the means remains significant.

Variations in systematic errors over time are a possibility that would influence the trend we calculate here. Examples include radiometric calibration and changes in the significance of interfering species because the reduced resolution microwindows are less well able to mask interfering lines from the retrieval. However, we note the F11 trend is similar between full and RR measurements (first pair vs. last pair). This would indicate that the effect of systematic errors has remained constant. More work is required to investigate this.

### 6.1 F11

A best-fit trend line for the equatorial atmospheres shows a change from 220 to 170 pptv in just over three years. The global mean trends, likewise, has the same value of 50 pptv.

This gives a trend of -15 pptv $yr^{-1}$, about -7.5 % $yr^{-1}$. The estimated error on the trend is about 10 pptv $yr^{-1}$, giving a trend range between -5 and -25 pptv $yr^{-1}$.

### 6.2 F12

A best-fit trend line for the equatorial atmospheres shows a change from 460 to 450pptv in just over three years. The global mean trends, likewise, has the same value of 10 pptv.

This gives a trend of -2.8 pptv $yr^{-1}$, about -0.6 % $yr^{-1}$. The estimated error on the trend is about 6 pptv $yr^{-1}$, giving a trend range between +4 and -8 pptv $yr^{-1}$.

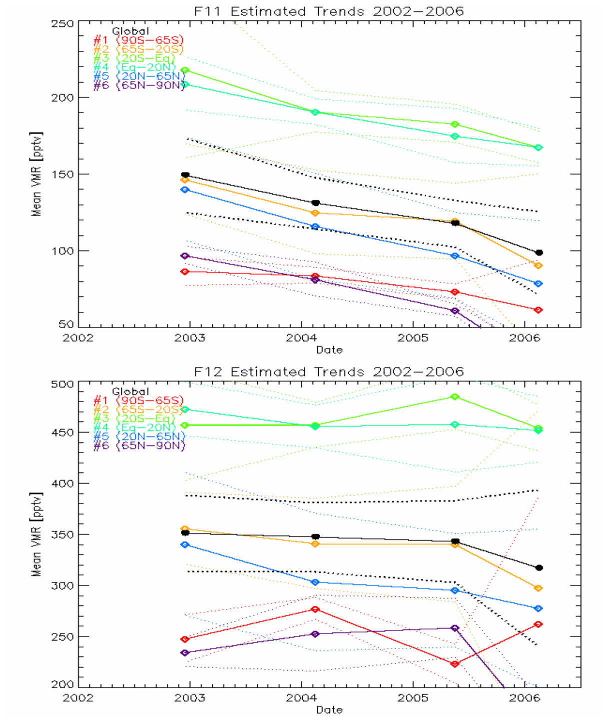


Figure 7: Estimated trends for F11 (top) and F12. Latitude bands are given by the differing colours (see legend). The global mean is shown in black. The standard deviations are shown as the dotted lines. Atmospheric variability will have influenced the size of the standard deviations recorded. The slight reduction in February 2006 values are not thought to be physical.

## 7. CONCLUSIONS

In this paper we hope to have demonstrated ‘operational’ reduced resolution retrievals from the new MIPAS measurement modes. This is very encouraging for the future as it demonstrates that even in the reduced mode; MIPAS is capable of good quality observations.

We have outlined how the microwindow error characteristics change, due to the loss of resolution of the lines of interfering species and the increase in random error through the reduced sampling time and vertical over-sampling.

The CFCs are a 'bonus' to the baseline mission expectations and are interesting scientifically.

We have estimated CFC-11 and CFC-12 trends as -7.5% (-15 +/- 10pptv $yr^{-1}$) and -0.6% $yr^{-1}$ (-2.8 +/- 6pptv $yr^{-1}$) respectively. The trends were obtained from global, height resolved fields making use of co-addition to improve random errors. These zonal mean fields contain useful information on the distribution of these species, their variability between different times of year, as well as the trend information discussed.

Our initial results are far from perfect, however. The F11 trend seems high when compared against surface measurements over a similar timescale. The trend from the surface measurements is at the edge of the estimated error on our trend measurement and is of the same sign. Significantly, the trend in F11 can be observed through measurements takes purely with full resolution data, allowing us to discount a number of possible systematic error terms that may have arisen through using the reduced resolution data and different scan patterns. Indeed, investigation of the practical effect of any major changes in systematic errors was one of the drivers behind this work.

### 7.1 Further work

The majority of the work will be in validation of the initial results presented here. For example, comparisons with other groups (initially full resolution comparisons only), [such as L. Hoffmann in these proceedings], will yield important information on the consistency of results from the same instrument but using different methods.

An outstanding validation of the operational reduced resolution retrieval is to degrade nominal resolution data to 0.0625 $cm^{-1}$ and compare resultant profiles with retrievals from the full resolution case.

Clearly, the use of more than three days in the month will reduce random error further. Likewise, adding other months will increase confidence in observed trends; or confirm our observation that the initial results give rise to a trend significantly larger than anticipated.

Finally, the region of the atmosphere over which the means and subsequent trends are calculated may benefit from a more robust separation into stratospheric and tropospheric air volumes. Large scale seasonal dynamical variability (such as descent at the winter pole) may also contribute to the trend we presented here. These effects need to be taken into account before a definitive result is possible.

For information on developments in our CFC work, see our website, www.atm.ox.ac.uk/group/mipas/. This work is not complete and the authors would welcome discussion as they continue their investigations.

### References

1. Blake, D., 2005, Chlorofluorocarbons (CFC-11, CFC-12, and CFC-113) in Whole-air Samples.

2. Dudhia A., Jay V., Rodgers C., 2002, Applied Optics, 41, 3665

3. Dudhia, A., 2002-2006, MORSE optimal estimation software. www.atm.ox.ac.uk/MORSE/