Measuring Halocarbons from the MIPAS instrument

University of Leicester

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INTRODUCTION

Halocarbons, such as CFC-11, CFC-12 and HCFC-22, are important trace constituents in the atmosphere through their role as greenhouse gases and their influence on stratospheric ozone chemistry. By using a limb sounding, spectrally resolving, instrument such as the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) onboard ENVISAT, with a resolution of 0.025 cm-1 unapodized, it is possible to distinguish the emission features of up to 30 trace species from the more densely packed spectral lines of the major constituent species such as CO_2 and H_2O. By utilising optimal estimation techniques, atmospheric concentrations of these trace gases can be obtained by inversion of the measured spectra. In nominal mode, the MIPAS measures emission spectra between 6 and 68 km, at approximately 3 km intervals, allowing profile information to be obtained. The work presented here focuses on a study using optimal estimation techniques to retrieve vertical profile concentrations of the halogen compounds CFC-11, CFC-12 and HCFC-22 from MIPAS level 1b spectral data.

MIPAS INSTRUMENT

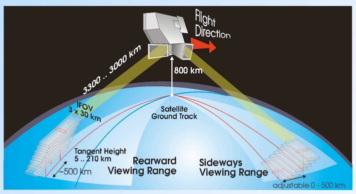

Fig 1. The limb viewing geometry of the MIPAS onboard ENVISAT

- Launched March 1st 2002 onboard ENVISAT
- Earth limb viewing Fourier Transform Spectrometer (FTS)
- Operates in the 685 cm^{-1} to 2410 cm^{-1} mid-infrared wavenumber range, resolution 0.025 cm^{-1} unapodized
- Performs routine elevation scans between 6-68 km in approximately 3 km steps, each of which takes around 4 seconds
- Continuous measurement capability due to Infrared source of radiation

RETRIEVALS FROM SIMULATED SPECTRA

In atmospheric spectroscopy, "**microwindows**" (MW's) are selected from the measured radiance spectra that are dedicated to target species and are **used for the retrieval of target gases**. This reduces computational costs and maximises efficiency in the retrieval.

To initially identify strong halocarbon emission regions, it is necessary to use a line by line forward model, here the **Oxford Reference Forward Model (RFM)**, to simulate atmospheric spectra based on known spectroscopic data and a reference climatology [Remedios,1999]. The ideal microwindow will have prominent emission lines of the target gas with only weak lines from any interfering species, i.e. gases that also emit in the same region of the spectrum.

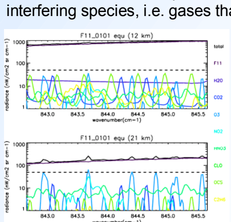

Figure 2. Simulated equatorial radiances of CFC-11 and important 'interfering' gases in the 842.650 – 845.650 cm-1 range (dotted line indicates the pre-flight estimate of the noise level for MIPAS band A)

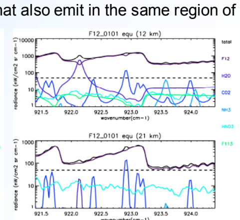

Figure 3. Simulated equatorial radiances of CFC-12 and important 'interfering' gases in the 921.4 – 924.4 cm-1 range (dotted line indicates the pre-flight estimate of the noise level for MIPAS band A)

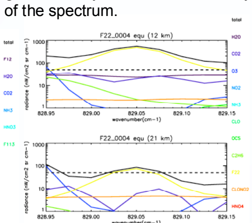

Figure 4. Simulated equatorial radiances of HCFC-22 and important 'interfering' gases in the 828.95 – 829.15 cm^{-1} range (dotted line indicates the pre-flight estimate of the noise level for MIPAS band A)

- **Strongest simulated emission features**:
  - **CFC-11 : 842.650 – 845.650 cm^{-1}**
  - **CFC-12 : 921.400 - 924.400 cm^{-1}**
  - **HCFC-22: 828.950 - 829.150 cm^{-1}**
- All three regions show calculated emission above the MIPAS noise equivalent spectral radiance of 50 nW/cm² sr cm^{-1}, at least between 12 and 21 km.
- Retrievals above 21 km likely to be influenced strongly by instrument noise

Leicester Retrieval Scheme:

- performs a joint retrieval by integrating spectral signals at each altitude in two distinct regions one of which is sensitive to the target gas, the other sensitive to aerosol
- in effect MIPAS data are treated as a radiometer rather than a spectrometer
- potentially advantageous for heavy halocarbons, where it is hard to distinguish individual line features

Scheme tested by firstly retrieving concentrations from simulated spectra (HCFC-22 results shown)

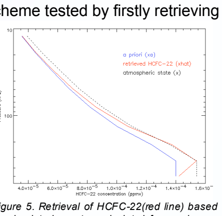

Figure 5. Retrieval of HCFC-22(red line) based on simulated spectra calculated from a known atmospheric state (black line), and starting the calculation from the a priori (blue line)

- Scheme reproduces the known 'measurements' very well between 80 and 130 mb
- Lower sensitivity to HCFC-22 at pressures below 80 mb and an increased bias towards our *a priori*

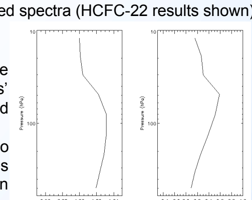

Figure 6. Residual spectra between 'measurements' based on simulated spectra and modelled radiances calculated by the RFM.

RETRIEVALS FROM MIPAS SPECTRA

Shown here are the results of running our retrieval scheme over MIPAS spectral data from **orbit 06117 on May 2nd 2003, for three different halocarbons (CFC-11, CFC-12 and HCFC-22)**. For CFC-11 and CFC-12, the Leicester retrievals are compared to retrievals performed by the University of Oxford's retrieval scheme (OPTIMO).

- No obvious systematic trend observable for either CFC-11 or CFC-12, compared to our climatology.
- Strong oscillations in some profiles, particularly in the polar summer case.
- Potentially an *a priori* bias at the South Pole due to poor measurement signal to noise.

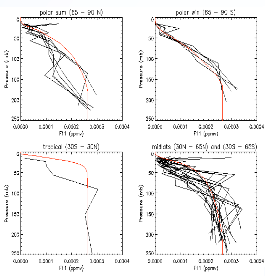

Figure 6. Retrieved CFC-11 profiles using Leicester scheme for orbit 06117 (black lines), compared to the a priori profile (red line)

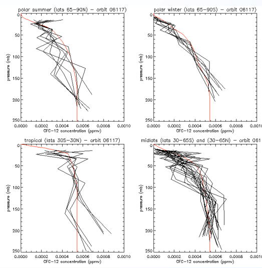

Figure 7. Retrieved CFC-12 profiles using Leicester scheme for orbit 06117 (black lines), compared to the a priori profile (red line)

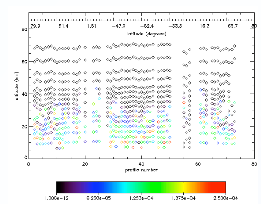

Figure 8. OPTIMO retrieved CFC-11 concentrations from orbit 06117 as a function of latitude

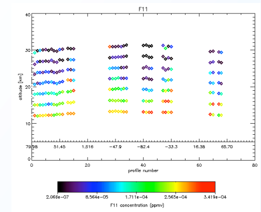

Figure 9. Leicester retrieved CFC-11 concentrations from orbit 06117 as a function of latitude

- Noisy CFC-11 measurements from both retrieval schemes
- No obvious zonal change in stratospheric CFC-11 concentration, compared to our climatology

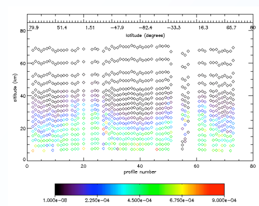

Figure 10. OPTIMO retrieved CFC-12 concentrations from orbit 06117 as a function of latitude

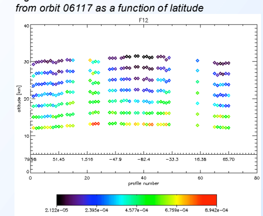

Figure 11. Leicester retrieved CFC-12 concentrations from orbit 06117 as a function of latitude

- Both schemes detect an apparent, strong, zonal variation of CFC-12 in the stratosphere, consistent with climatology,

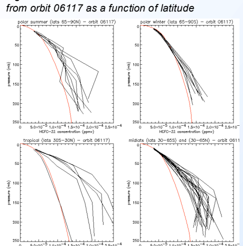

Figure 12. Retrieved HCFC-11 profiles using Leicester scheme for orbit 06117 (black lines), compared to the a priori profile (red line)

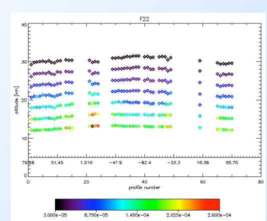

Figure 13. Leicester retrieved HCFC-22 concentration from orbit 06117 as a function of latitude

- Enhancement of HCFC-22 concentrations, compared to climatology, in all four regions.
- Effect most pronounced in the tropics and mid-latitudes.
- Smaller increase towards high latitudes

DISCUSSION

- Retrievals of three chemically and radiatively important halocarbons (CFC-11, CFC-12 and HCFC-22) from the MIPAS orbit 06117 are presented. **Results are preliminary and should be treated with caution.**
- HCFC-22 retrievals from simulated spectra represented the upper troposphere and lower stratosphere region particularly well but showed a systematic bias towards our *a priori* climatology at pressures below 80 hPa.
- Potentially a global enhancement of HCFC-22 compared to our climatology.
- Little variation between climatology and retrieved CFC-11 and CFC-12 concentrations. Result consistent between both retrieval schemes. However, a potential CFC-11 and CFC-12 decrease at the north pole.
- **A full error analysis on these preliminary results is currently in progress.**

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