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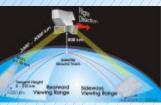
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INTRODUCTION

...chlorofluorocarbon-22 (HCFC-22) is an important trace constituent in the atmosphere ...th its role as a greenhouse gas and its potential influence on stratospheric ozone chemistry. ...t estimates of its vertical distribution are limited to infrequent and sparse balloon and ...n campaigns, giving poor spatial coverage. In particular, knowledge of its global vertical ...variation is poor but could be improved by utilising satellite remote sensing techniques.

...ng limb infrared sounding, spectrally resolving, instruments such as a Fourier Transform ...ometer (FTS) it is possible to measure the infrared emission of radiation from trace gases ...nce obtain atmospheric concentrations by inversion of the measured spectra. A recent ...ment has been the launch of the Michelson Interferometer for Passive Atmospheric ...ng (MIPAS) onboard the ENVISAT satellite. The work presented here focuses on the ...d study of the spectral signatures of HCFC-22 in MIPAS level 1b data and on the potential ...cal profile retrievals from the MIPAS instrument.

...PAS INSTRUMENT

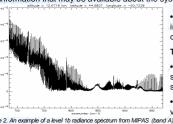*The limb viewing geometry of the MIPAS onboard ENVISAT*

- 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

...TIMAL ESTIMATION

...evals from remotely sensed radiance measurements from the limb sounding MIPAS ...ment, or the *inverse problem,* involves finding the best representation of the atmospheric ...entrations of 'target' gases given the measurements made, together with any appropriate ...information that may be available about the system and the measuring device.

...e 2. An example of a level 1b radiance spectrum from MIPAS (band A)

- The radiance spectrum contains spectral information on atmospheric trace gas concentrations

Two questions arise:

- How is it possible to distinguish which spectral lines belong to which atmospheric species?
- How can we convert these radiances into volume mixing ratios of gases?

...ciated with the inverse problem is the required understanding and description of the ...mation content of the measurement, the relationship between the true state of the system ...hat retrieved using inverse methods, the error analysis of the overall measuring system, ...isation of the observing systems, and validation of the results.

...e are various techniques available to estimate the atmospheric concentration of a target ...from limb radiance measurements, the one to be utilised for this study and hence described ...is *optimal estimation* (Rodgers, 1976). This technique constrains the profile to be close to ...a *priori* solution **a**, covariance S_a and minimizes the cost function c:

$$c^2 = (y - Kx)^T S_y^{-1}(y - Kx) + (x - a)^T S_a^{-1}(x - a) \quad (1.1)$$

...e **y** is the measurement, **K** is the Jacobian, **x** is state of the atmosphere and S_y the ...ance on the measurements.

...also has a 'regularising' effect by smoothing the differences between **x** and **a** through ...ve correlations in S_a

$$a = G(y - Ka) \quad (1.2)$$

$$...re\ G = (K^T S_y^{-1} K + S_a^{-1})^{-1} K^T S_y^{-1} \quad (1.3)$$

$$= (K^T S_y^{-1} K + S_a^{-1})^{-1} \quad (1.4)$$

Note: as $S_a \to \infty$, converges to a Least Squares Fit type solution

...advantage is that solutions exist for complete range of measurements m = 0 ..n .. (not just ...where n is the number of state parameters]) so

...ible to always get a profile even if measurements are missing

...uential retrieval with a subset of measurements

...isadvantage of this technique is that *a priori* introduces bias into the estimate

$$\hat{x} = Ax + (I - A)a \quad (1.5)$$

...e the *averaging kernal*, **A = GK**. So care has to be taken when averaging profiles!

MICROWINDOW STUDIES

To identify regions of the mid-infrared spectrum where HCFC-22 is radiatively active wit... Jacobian spectrum (1 % perturbation) for 12 km tangent altitude was compared to real M... spectra for a comparable altitude. This also identifies the spectral region from which HCF... can be retrieved. (All simulations performed using the Oxford forward model, RFM).
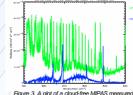

Figure 3. A plot of a cloud-free MIPAS measured spectrum and the jacobian of HCFC22

- Simulated spectral signatures of HCFC-22 are cross-sections nature with sharp Q branches
- Sharpest HCFC-22 features at 809 cm^1 and 829...
- Spectral signature of HCFC-22, in the 785 – 840... range, is masked by stronger lines from... atmospherically abundant gases with signatures... region such as CO_2 and H_2O.

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

Selecting regions where the HCFC-22 peak is strongest in the simulated spectra and calc... the expected radiance contribution from each "interfering" gas defines smaller suitable MW's...

- Only the region 828.95 – 829.15 cm^1 (right) has a dominating simulated radiance contribution from HCFC-22
- Simulating the limb radiance in this region shows that HCFC-22 may be detectable from MIPAS above the pre-flight expected noise equivalent spectral radiance of 50 nW in band A, at least between 12 and 21 km
- The region may become unsuitable for retrievals above 21 km as the simulations suggest that the HCFC-22 signal will fall below the noise level.

Figure 4. Simulated equatorial radiances of HCFG22 and important I... gases in the 828.95 – 829.15 cm-1 range (dotted line indicates the p... estimate of the noise level for MIPAS band A)

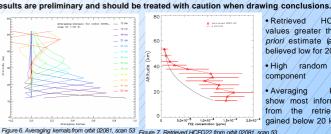To determine if there is a relationship betwee... simulated spectrum in figure 4 and a measured... spectrum, the cloud-free correlation coefficients... been calculated over latitudes for one orbit (020...

- $r \geq 0.8$ over all global latitudes
- Slightly lower coefficients towards Antarctica
- Using student ttest to determine significance... km finds relationship around the 95 % confidence...

Figure 5. Cloud-free correlation coefficients calculated for MIPAS orbit 02081 between MIPAS spectra and the 12 km HCFC-22 jacobian

VERTICAL HCFC-22 RETRIEVALS

This initial study into the possibility of retrieving HCFC-22 from MIPAS has utilised cloud-free... **1b radiance spectra from orbit 02081 (Jul. 2002).** An optimal estimation retrieval has... performed for scan 53 of the orbit and the results are shown below. **It should be noted that... results are preliminary and should be treated with caution when drawing conclusions.**

Figure 6. Averaging kernals from orbit 02081, scan 53

Figure 7. Retrieved HCFC22 from orbit 02081, scan 53

- Retrieved HC... values greater th... *priori* estimate (a... believed low for 20...
- High random component
- Averaging k... show most infor... from the retrie... gained below 20 k...

- Very encouraging signs that it is possible to retrieve profiles of HCFC-22 from MIPAS usin... 828.95 – 829.15 cm^1 region of the spectrum.
- This retrieval reproduces the increasing atmospheric concentration trend of HCFC-2... highlighted in the latest IPCC report (IPCC, 2001)
- This will be extended to other regions of the world to test feasibility of global profile retrieva...
- Other microwindows that have been identified to contain HCFC-22 spectral information w... utilised in future retrievals to enhance information content of the spectra.

...ences: IPCC, 2001: Climate Change 2001: The Scientific Basis. Contribution of Working Group 1 to the Third...