ENVISAT MIPAS MEASUREMENTS OF CFC-11 AND CFC-12: AN ADD-ON TO ESA OPERATIONAL DATA

L. Hoffmann, R. Spang, M. Kaufmann, and M. Riese

Forschungszentrum Jülich, ICG-I, 52425 Jülich, Germany

Abstract

Envisat MIPAS measurements cover nearly continuously the period July 2002 to March 2004. ESA offline Level 1 and Level 2 reprocessing of MIPAS measurements during this period is completed by now. Calibrated infrared radiance spectra (4 – 15 μm) and atmospheric data (vertical profiles of pressure, temperature, and six trace gases) are available for further analysis. A general fast forward model and an optimal estimation retrieval processor suited to derive additional trace species from MIPAS measurements as well as other mid-infrared limb-sounding experiments have been developed in Juelich. The retrieval system was applied to derive the global distributions of CFC-11 and CFC-12 abundance and aerosol extinction coefficients near 10.7 μm and 12.0 μm from the MIPAS radiance measurements. Error studies indicate good quality of the retrieved CFC-11 and CFC-12 data (total error 4 – 7 %). Further studies show that the influence of a priori on the obtained results is rather small (below 5 – 10 % at most heights). Comparisons against MIPAS retrievals carried out by other working groups and first successful validations with in situ measurements show that the retrieved data are suited for further scientific analysis. Climatological mean values and standard deviations compare well against results of other satellite experiments.

Key words: Envisat; MIPAS; chlorofluorocarbons; CFC-11; CFC-12.

1. INTRODUCTION

Monitoring the global distribution of chlorofluorocarbons (CFCs) is very important for several reasons: (1) The chlorine released by photolytic decomposition of CFCs plays a major role in catalytic ozone destruction in polar stratospheric chemistry. (2) CFCs are strong infrared absorbers. They contribute significantly to the anthropogenic greenhouse effect. (3) Being long-lived trace species, CFCs are well suited to observe and quantify transport of air masses in the upper troposphere and lower stratosphere.

The Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) instrument [1, 2] aboard ESA’s Envisat can be used to monitor the global distributions of CFCs. MIPAS measures the infrared emissions of atmospheric constituents arising at the atmospheric limb. Radiance measurements cover the range 4 – 15 μm with a high spectral resolution of about 0.025 cm⁻¹. Vertical sampling of the atmospheric limb covers the altitude range 6 – 68 km with a sampling step of 3 km, corresponding to the vertical field-of-view of the instrument.

Strong infrared emissions of CFC-11 (CFCl₃) and CFC-12 (CF₂Cl₂) are present near 850 cm⁻¹ and 920 cm⁻¹, respectively. Fig. 1 shows parts of simulated MIPAS spectra for 9 km and 15 km tangent height at mid-latitude climatological conditions. Gray shaded areas indicate spectral ranges used for the joint retrieval of either CFC-11 or CFC-12 and aerosol extinction coefficients. Optimal spectral ranges were found by a detailed analysis of Shannon information content which aims on minimizing the total retrieval error compared to a priori uncertainties.

2. A FAST FORWARD MODEL FOR ENVISAT MIPAS

A new fast forward model suitable for interpretation of measurements made by Envisat MIPAS and other mid-infrared remote-sensing experiments was developed recently as part of a PhD thesis in our group [3]. The Juelich Rapid Spectral
Figure 1. Simulated Envisat MIPAS spectra for a) 9 km and b) 15 km tangent height and mid-latitude mean atmospheric conditions. Shown are total limb radiance as well as radiance contributions due to CFC-11, CFC-12, and aerosols.

Simulation Code (JURASSIC) can be used in any case where scattering is negligible (i.e., for cloud-free conditions in the mid-infrared spectral region) and where local-thermodynamic equilibrium applies (i.e., in the troposphere and stratosphere). The model is most flexible with respect to specification of observation geometry (observer inside or outside the atmosphere; limb-, nadir-, or up-looking-observations) and specification of atmospheric data (homogeneously stratified atmosphere for comparison against other models and for conventional retrievals; 4D data in form of ensembles of air parcels for studies of radiative transfer, more complex retrieval schemes, or data assimilation).

Rapid radiative transfer calculations are facilitated by means of the emissivity growth approximation (EGA) [4, 5]. Radiative transfer is calculated approximately based on spectral mean values rather than the exact line-by-line approach. Spectral mean emissivities are obtained by simple and fast interpolation from precomputed look-up tables. The EGA method and the use of emissivity look-up tables allows to reduce the CPU-time required by JURASSIC by 2–5 orders of magnitude compared to line-by-line forward models. The errors in the radiative transfer calculations due to the EGA are small (generally below 1–2%) and often negligible compared to other error source (e.g., errors caused by uncertainties of spectroscopic data). As an example, Fig. 2 shows a comparison of mean radiances at the center of the 11.8 μm band of CFC-11 computed by means of JURASSIC and the MIPAS Reference Forward Model (RFM) [6]. The deviations found in the stratosphere and upper troposphere are below 0.5% for all climatological cases.

Figure 2. Comparison of mean radiances (844 – 851 cm⁻¹) in the 11.8 μm band of CFC-11 based on forward calculations using the RFM (line-by-line calculations) and JURASSIC (EGA method). Comparisons are done for different climatological conditions (see legend). In tropical latitudes the band gets optically thick below 8 km tangent height.
3. RETRIEVAL OF CFC-11 AND CFC-12

Retrieval of CFC volume mixing ratios and aerosol extinction coefficients from Envisat MIPAS radiance measurements is based on the standard optimal estimation approach [7]. The best estimate of the atmospheric state is determined by minimizing the differences between the MIPAS radiance measurements and simulated observations, provided by the forward model for a given estimate, as well as minimizing the differences between the estimate and the a priori state. The minimization problem is solved straightforward by means of the Levenberg-Marquardt method. Some technical issues are discussed in [8, 9].

Fig. 3a shows the results of the detailed error analysis for a typical CFC-11 retrieval. The total error is in the order of 9 – 13 ppt (4 – 7 %). Leading retrieval errors in the upper part of the retrieved CFC-11 volume mixing ratio profile are due to noise (up to 7 – 8 ppt), errors of offset calibration (7 – 8 ppt), and uncertainties due to the spectral extrapolation of aerosol extinction coefficients (5 – 6 ppt). Leading retrieval errors in the lower part of the profile are due to errors of gain calibration (8 – 9 ppt), uncertainties of temperature (5 – 6 ppt), CFC-11 spectroscopic data (4 – 5 ppt), and pressure (3 – 4 ppt). Retrieval errors caused by the use of approximated radiative transfer calculations and due to interfering species are of minor importance (1 – 3 ppt). Error analyzes for CFC-12 show similar results.

Even if error analysis like in Fig. 3a are frequently shown, it must be taken into account that the results comprise some major uncertainties since information about temporal, spatial or spectral correlations of instrument errors, parameter errors, or forward model errors is missing. In our retrieval a first order auto-regressive model is applied to model these correlations. Correlation lengths are used to describe the scales on which an error is a statistic or systematic one. As an example, Fig. 3b shows the influence of vertical correlation length $c_z$ of offset calibration errors on the estimated retrieval error. Varying between uncorrelated ($c_z = 0.1$ km) and fully correlated ($c_z = 10000$ km), the retrieval error may either be dominating or negligible at certain heights. As no further information about the correlation length of offset calibration errors is available, an educated guess had to be made. First assumption would be to use full correlations for a single profile as on-board calibration is carried out after every fourth vertical scan. On the other hand, stray-light effects or temperature drifts probably lead to reduced correlation lengths. Hence, the guess $c_z = 10$ km might be reasonable.

To analyze the influence of a priori data on the retrieval results the smoothing error was estimated. Fig. 3c shows the results for a typical retrieval. To estimate the smoothing error the covariance matrix of a real ensemble of atmospheric states must be known. In our case the a priori covariance was used as ensemble covariance. Diagonal elements of the a priori covariance matrix were obtained from a climatology [10]. Fig. 3c shows the influence of a scaling factor for the climatological standard deviations. Evidence suggests that the original standard deviations provided by the climatology are too small. Hence, a scaling factor of 300 % was applied during the retrieval process. The estimated smoothing error is about 5 – 10 ppt. Off-diagonal elements of the a priori covariance were determined by a first order auto-regressive model with a vertical correlation length of 10 km. In case of uncorrelated a priori data the smoothing error would shrink to 3 – 6 ppt, but retrieval oscillations occur. Introducing a correlation length of 10 km efficiently reduces retrieval oscillations and causes a reduction of vertical resolution of the observations from about 3 km to 4 km, only.

4. VALIDATION AND ANALYSES OF CFC MEASUREMENTS

Several cross-checks and validation activities were carried out to improve and ensure the quality of the derived CFC-11 and CFC-12 datasets. Primarily to check the forward model and the accompanying retrieval processor, comparisons were carried out between our retrievals and results obtained at Forschungszentrum Karlsruhe, IMK. The IMK retrievals are based on exact line-by-line forward calculations [11]. Scatter plots for individual days show good agreement between the results. For CFC-11 a high-bias of 8 % was detected in our data which is caused by different versions of spectroscopic data and will be removed in further data versions. For CFC-12 the bias between the results is below 1 %. Scattering observed between individual profiles is mainly due to different temperature data, on which the CFC retrievals react very sensitive. Temperature deviations in the order of 1 – 3 K were found between ESA data [12, 13], on which our retrievals are based on, and independently derived IMK temperatures [14]. A minor part of scattering is due to different versions of Level-1B data (near-real-time vs. off-line data).

Fig. 4 shows a comparison between climatological mean profiles for CFC-11 and CFC-12 derived from our Envisat MIPAS retrievals and measurements made from space by other remote-sensing experiments [15–19]. In this comparison only mid-latitudes observations (20° – 65° N/S) are considered. Good agreement between the different experiments is found. The shape of the vertical profiles agrees very well. In most cases only a small constant offset between the mean profiles is present which may be caused by systematic errors or trends. ATMOS (Atmospheric Trace Molecule Spectroscopy) measurements have a good accuracy (errors well below 5 %). The mean profiles, however, show a larger variability since the number of measurements is rather small. The CRISTA-2 (Cryogenic Infrared Spectrometers and
Telescopes for the Atmosphere) CFC-11 profile corresponds very well to the MIPAS data. For CRISTA-1 a steeper gradient is observed. Retrievals for CRISTA-1 are more complicated due to an enhanced aerosol background caused by the volcano eruption of mount Pinatubo in 1992. This might also be the reason why large deviations are found for CFC-11 between the climatology (derived from Cryogenic Limb Array Etalon Spectrometer (CLAES) measurements in 1991 – 1993) and the other datasets. The effect of the misleading climatology on the MIPAS retrievals is small since climatological standard deviations were increased by 300 %, allowing for a greater variance of atmospheric conditions.

Fig. 5 shows time series of the CFC-11 volume mixing ratio zonal mean and standard deviation at 20 km altitude. The period June 2002 to March 2004 is nearly continuously covered by MIPAS measurements. To derive the time series a set of 430,000 Envisat MIPAS vertical profiles was analyzed. A seasonal interdependence is found in the zonal means (Fig. 5a). Higher CFC-11 volume mixing ratios occur in the summer hemisphere. More clearly the seasonal interdependence is to be observed in the standard deviations (Fig. 5b). In tropical latitudes (depending on season between 10°S – 20°N or 20°S – 10°N) the standard deviations are below 5 – 10 ppt. At mid and high latitudes standard deviations up to 50 ppt are found. At mid and high latitudes short periods (3 – 5 month) of significantly reduced variability are to be observed in late summer and early autumn. On the southern hemisphere highest standard deviations are found near 60°S. On the northern hemisphere the regions of high standard deviations are less concentrated on a certain latitude band. This behavior originates from atmosphere dynamics. In the antarctic winter a very stable polar vortex exists. In the arctic winter the polar vortex is disturbed e. g. by orographically excited planetary waves. In the southern hemisphere small shifts of the polar vortex (planetary wave-1 events) lead to the observed small band of high standard deviations. The northern polar vortex is deformed in a more complex manner (planetary wave-2 events). The polar region is less restricted from mid latitudes here. The enhanced large-scale mixing of CFC-11 poor polar air masses and mid latitude air masses with high CFC-11 concentrations causes the observed spatial distribution of high variance over a rather wide latitude range.

To facilitate a more detailed analysis of the derived MIPAS CFC-11 and CFC-12 data sets a Kalman filter module was recently developed for the Chemical Lagrangian Model of the Stratosphere (CLaMS). The filter is especially suited for
validation purposes. First comparison between assimilated MIPAS CFC-11 and CFC-12 measurements and corresponding data obtained during Envisat MIPAS validation campaigns by the the in-situ High Altitude Gas Analyzer (HAGAR) instrument aboard the M-55 Geophysica aircraft show a good consistency. First tests with the Kalman filter also suggest that the internal consistency of the derived datasets is even better than predicted by the error analysis.

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REFERENCES


