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MARC: A code for the retrieval of atmospheric parameters from millimeter-wave limb measurements

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Abstract

A new data analysis software is presented that has been developed for the retrieval of atmospheric minor constituents from limb-sounding observations made in the millimeter and sub-millimeter spectral regions. The code, which is called MARC (Millimetre-wave Atmospheric-Retrieval Code), has been designed to analyze the observations of the MARSCHALS (Millimetre-wave Airborne Receivers for Spectroscopic CHaracterisation in Atmospheric Limb-Sounding) instrument which operates on the M-55 stratospheric aircraft. The main objective of the analysis of MARSCHALS observations will be to assess long-wave measurement capabilities for the study of the upper troposphere and lower stratosphere regions. The key questions will be the accuracy and spatial resolution that can be achieved by long-wave measurements in presence of clouds and horizontal gradients.

MARC performs a global-fit multi-target retrieval, in which optimal estimation is used and errors of the forward model parameters are taken into account for the definition of the cost function minimized in the retrieval. With these features it is easy to use the variables of the problem as either forward model constant parameters or retrieved unknowns with minimum impact on the stability of the retrieval. MARC can perform a wide spectral-band analysis of the observations without a selection of the analyzed channels, and the retrieval process provides an error budget of the retrieved unknowns that includes both the forward model errors and the measurement errors. The error budget obtained in this way is smaller than that obtained when accounting a posteriori for the systematic errors. The new combination of the retrieval features makes possible an efficient and optimal exploitation of the information content of the observations.

Keywords: Remote sensing; Limb-sounding; Retrieval techniques; Heterodyne instruments; Atmosphere

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1. Introduction

The determination of the vertical distribution of atmospheric parameters, such as pressure, temperature and minor constituents, from remote sensing observations is a major technological achievement and a unique investigation tool for the study of atmospheric chemical and dynamical processes.

For this purpose, in recent years, several space-borne limb-sounding spectrometers have been deployed. These instruments provide a much larger number of spectroscopic observations than what was achieved with previous limb sounding or nadir sounding radiometers. This is in particular the case of Fourier transform instruments. For example the MIPAS (Michelson Interferometer for Passive Atmospheric Sounding) [1,2] instrument, which flies on the Envisat platform of ESA (European Space Agency), combines the wide spectral range and high spectral resolution of a Fourier Transform Spectrometer (FTS) with the continuous measurements of the emission limb-sounding technique for the observation of a large number of chemical species with an extensive geographical coverage. ILAS (Improved Limb Atmospheric Spectrometer) [3] and ACE (Atmospheric Chemistry Experiment) [4,5] are also FTS limb-sounding instruments which observe a large number of chemical species using the solar occultation technique.

The emission limb-sounding technique is also used in the case of the SMR (Sub-Millimetre Radiometer) instrument which operates on board of the Odin satellite [6], and of MLS (Microwave Limb Sounder) [7] instrument, which flew on the UARS (Upper Atmosphere Research Satellite) platform. Both SMR and MLS are microwave spectrometers which combine the emission limb-sounding technique with spectroscopic measurements at high spectral resolution with limited spectral coverage. With the recent launch by NASA (National Aeronautics and Space Administration) of the Aura platform, a few more instruments are operative, such as HIRDLS (High Resolution Dynamics Limb Sounder) [8], TES (Tropospheric Emission Spectrometer) [9] and the new MLS.

All the codes used to retrieve the atmospheric parameters from these observations operate with an inversion procedure in which the forward model (FM), $\mathbf{F}(\mathbf{x}, \mathbf{b})$, which describes the observations $\mathbf{y}$, is inverted, and the unknown $\mathbf{x}$ are determined given the estimates $\mathbf{b}$ of the known parameters (atmospheric, instrumental and geometrical parameters). The inversion is made with an iterative procedure that provides the minimization of a cost function. Despite the common procedure of the retrieval and the common features of these instruments, which all provide hyperspectral limb-sounding measurements, the different operating modes, spectral regions and spectral resolutions have led to the development of several codes.

Retrieval codes evolve either with improved accuracy in the FM calculations or with the adoption of more comprehensive inversion approaches. Examples of the first type of improvement are the modeling of non-local thermodynamic equilibrium (NLTE) of several atmospheric constituents (see, e.g. [10,11]) and the modeling of CO_2 line mixing [12]. Several examples can be recalled of the second type of evolution, which follows the continuous improvement of computer performances. Park and Carli [13] performed the simultaneous retrieval of both atmospheric and instrument variables. Carlotti [14] replaced the classical onion-peeling technique [15,16], in which the individual limb views are sequentially analyzed, with the global-fit of the whole sequence of limb observations. In recent years, significant efforts have also been made for the development of two-dimensional retrieval codes in which a set of limb sequences is simultaneously analyzed for the retrieval of a two dimensional field of the atmospheric target unknowns [17–20]. This approach enables a proper modeling of the horizontal variability of the atmosphere, and often makes it possible to improve the usually poor horizontal resolution of limb-measurements [20,21]. The retrieval problem can be made more complex, not only by increasing the number of locations in which the retrieval is performed, but also by increasing the number of retrieved unknowns in each location. The first type of retrieval is usually referred to as single-target retrieval and the second as multi-target retrieval [22,23]. Indeed, Carlotti et al. [24] have recently presented an open source code which combines two-dimensional and multi-target capabilities. A further feature that may characterize a retrieval code is the approach used to account for the FM errors. For instance, the retrieval code used for the analysis of MLS measurements [25] adds to the error covariance of the radiances an additional term, which accounts for the uncertainties of the FM errors due to the interfering species.

The deployment of a new limb sounding instrument, called MARSCHALS (Millimetre-wave Airborne Receivers for Spectroscopic CHaracterisation in Atmospheric Limb-Sounding), has identified new retrieval requirements which called for the development of a new dedicated code. The Millimetre-wave

Atmospheric-Retrieval Code (MARC) that was developed for the analysis of MARSCHALS measurements is discussed in this paper.

2. The MARSCHALS instrument

MARSCHALS [26] is a heterodyne limb-sounding spectrometer that measures the atmospheric thermal emission in the millimeter and sub-millimeter wave regions and that operates on board the M-55 stratospheric aircraft (Geophysica) which can fly at altitudes up to 21 km. MARSCHALS is an ESA instrument and it has been developed as a simulator of the MASTER (Millimetre-wave Acquisition for Stratosphere–Troposphere Exchange Research) instrument [27], which is proposed for future Earth Explorer Missions.

The objective of MASTER is the study of the Upper-Troposphere and Lower-Stratosphere (UTLS), and the instrument is designed to operate in a spectral region where cloud attenuation does not make the atmosphere opaque and minor atmospheric constituents can be observed for the identification of chemical and dynamical processes in the presence of clouds. These measurement capabilities of the millimeter and sub-millimeter wave regions have not been demonstrated yet with actual instruments, and the difficulties that may be posed by water vapor attenuation, horizontal gradients and cloud properties must be quantified [28]. The MARSCHALS instrument was developed to study the scientific capabilities of this unexplored spectral region with real observations in order to analyze the extent of its advantages and to validate the spectro-radiometric requirements of MASTER.

MARSCHALS observes the limb emission of the atmosphere in three bands centered around 300, 320 and 345 GHz. These bands correspond, respectively, to band B, C and D of the MASTER instrument and for reasons of continuity they keep the same names also in MARSCHALS.

The incoming radiation is split into the three bands by a quasi-optical network, and is coupled into the individual front-end mixers. The image side-bands are filtered out. Each band has a band-width of 12 GHz, which is split into a number of sub-bands by a second down-conversion stage in order to be compatible with spectrometer technology. The spectrometers measure the spectral power density with a resolution of 200 MHz, and the resulting digitized outputs are available for ground processing.

The atmosphere is observed with a 220 mm diameter scanning antenna and the pointing is obtained with a drive mechanism that allows elevation angles from -5° to $+20^\circ$. When MARSCHALS instrument operates on the Geophysica aircraft, a scan through the limb can be made from the Earth's surface to platform altitude (about 21 km) with additional elevation angles above the horizontal for a view of the sky with minimum atmospheric signals. The root mean square (rms) beam pointing knowledge during scans is better than 0.0025°. The field of view of the antenna is approximately 0.34° in both the horizontal and vertical directions. The calibration switching network consists of a hot calibration load with a nominal radiometric temperature of about 300 K and a cold calibration load with a nominal radiometric temperature of about 88 K. The instrument specifications require a radiometric error characterized by a NET (Noise Equivalent Temperature) of less than 1.5 K, with the goal of 1 K.

The targets measurements of MARSCHALS are H_2O, O_3, HNO_3, N_2O, CO, and temperature profiles in the UTLS. Fig. 1 shows simulations of the spectra that will be measured by MARSCHALS. More detailed information on the MARSCHALS instrument can be found in [26].

3. MARC retrieval code

MARC is the code developed for the Level-2 analysis of MARSCHALS measurements and will become the ESA tool for MARSCHALS data processing. In Level-2 analysis, the calibrated and geolocated spectra measured by the instrument are fitted with a FM calculation for the retrieval of the vertical profiles of the target quantities.

An accuracy better than 10% and a vertical resolution better than 2 km [28] are desirable in the determination of the volume mixing ratio (VMR) of the target species in order to detect the discontinuities that characterize UTLS processes. These objectives are difficult to achieve, and may not be equally feasible for all target species at all altitudes.

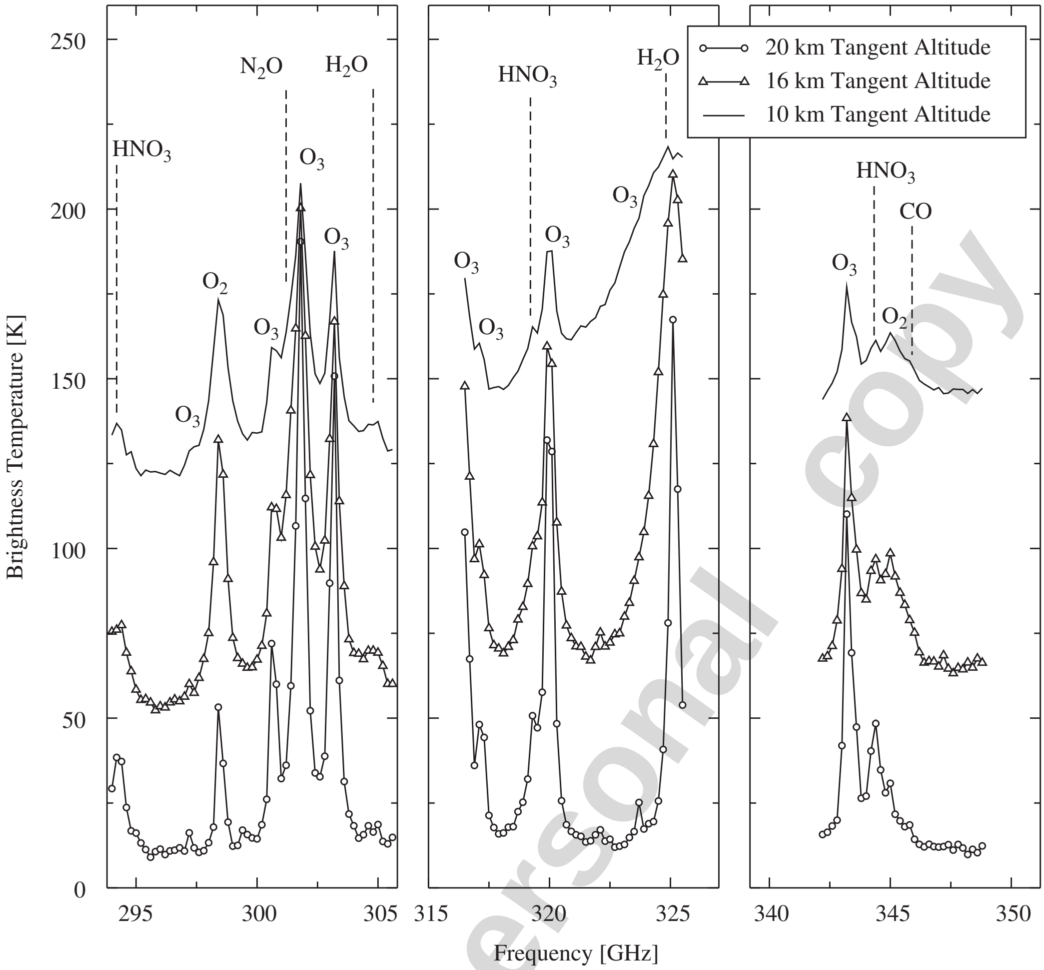

Fig. 1. Simulated sub-millimeter spectra of the atmospheric emission in the three bands of MARSHALS at different tangent altitudes. The signatures of the main target species are labeled. A NET of 1 K (rms) has been added to the spectra.

This analysis is similar to that performed with the ORM (optimized retrieval model) code [29], which was developed for the operational analysis of the MIPAS instrument. Several solutions adopted in that code have also been applied to MARC. This is, in particular, the case for the FM calculation for which a similar baseline has been adopted despite some upgrades for the modeling of clouds and horizontal gradients as well as a different modeling for the line-shape, the continuum emission and the refractive index for the different spectral region. On the other hand significant differences exist in the retrieval approach.

Compared to FTS observations, MARSCHALS measures relatively few spectral channels (about 150 channels for each limb-angle, instead of the 10^5 channels of MIPAS), and several unknowns (the target quantities) and FM errors (the errors in the assumed FM parameters) simultaneously affect these channels. These experimental circumstances imply the use of a different retrieval approach.

In the case of MIPAS, it is possible to identify for each target species a sub-set of useful spectral channels, in which the contribution of the target quantity is maximal and the interference of errors is minimal. The small spectral intervals which contain the most useful spectral channels are called *micro-windows* [30,31]. The retrieval is performed sequentially for the individual targets, and a few selected micro-windows are analyzed for each of them. Therefore, the MIPAS retrieval is a sequential single-target retrieval with micro-window selection.

In the case of MARSCHALS, the small number of measured spectral channels can be simultaneously analyzed and it is possible to perform a multi-target retrieval with no micro-window selection (wide-band retrieval). This approach has the advantage of avoiding the channel selection and of exploiting all the available data. The disadvantage is that the FM errors can no longer be considered as negligible and have to be accounted for in the retrieval. This means that FM errors must be evaluated together with the measurement error when the cost function of the retrieval is calculated. Wide-band retrieval also implies that channels are analyzed in which the contribution of some instrumental parameters is comparable to that of the retrieval targets. These instrumental parameters are observed with a sensitivity that makes them eligible for the retrieval. A smooth transition from FM parameter to retrieved unknown can be obtained using the optimal estimation (OE) method [32].

On the basis of these considerations, MARC is designed as a global-fit, multi-target retrieval, in which FM errors are accounted for in the inversion process and OE is used. The code can be used for a wide-band analysis of the observations and accounts for horizontal gradients and cloud contamination.

These choices of MARC are individually discussed in the following sections, in the frame of the mathematics of the retrieval briefly recalled in the next section.

4. Mathematics of the retrieval

The mathematics of inverse geophysical problems has been discussed in detail by Rodgers [32]. The main relevant equations are here recalled for a more clear characterization of the MARC choices.

The FM simulates the observations $\mathbf{y}$. However, even a perfect FM cannot reproduce exactly the observations due to the measurement noise ε. Observations and FM simulations are therefore linked by the following expression:

$$\mathbf{y} = \mathbf{F}(\mathbf{x}, \mathbf{b}) + \varepsilon, \tag{1}$$

where $\mathbf{x}$ is a vector containing the retrieval unknowns that are the objective of the measurement and $\mathbf{b}$ is a vector containing the FM parameters which are not the objective of the retrieval and for which a reasonable estimate exists. Each of the quantities involved ($\mathbf{y}$, $\mathbf{x}$ and $\mathbf{b}$) may have more than one dimension (for instance, spectral and tangent altitude dependence in the case of observations made using the limb-sounding technique), however, it is sequentially organized in a one-dimensional vector.

The retrieval involves the determination of the $\hat{\mathbf{x}}$ vector that minimizes the cost function. In the unconstrained non-linear least-square fit (NLSF) approach, the cost function is equal to

$$(\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}}, \hat{\mathbf{b}}))^{\mathrm{T}} \mathbf{S}_T^{-1} (\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}}, \hat{\mathbf{b}})), \tag{2}$$

where $\mathbf{S}_T$ is the variance–covariance matrix (VCM) of the residuals $(\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}}, \hat{\mathbf{b}}))$ of the fit and $\hat{\mathbf{b}}$ are our estimates of the FM parameters. Two error sources affect the residuals: error ε, due to the measurement noise of the $\mathbf{y}$ observations, and the uncertainties ε_b of $\hat{\mathbf{b}}$. Therefore

$$\mathbf{S}_T = \mathbf{S}_y + \mathbf{S}_{\mathrm{FM}}, \tag{3}$$

where $\mathbf{S}_y$ is the VCM of the measurement errors, equal to

$$\mathbf{S}_y = \langle \varepsilon \varepsilon^{\mathrm{T}} \rangle \tag{4}$$

and $\mathbf{S}_{\mathrm{FM}}$ is the VCM of the errors in the estimates of the FM parameters, equal to

$$\mathbf{S}_{\mathrm{FM}} = \sum_b \langle (\mathbf{K}_b \varepsilon_b)(\mathbf{K}_b \varepsilon_b)^{\mathrm{T}} \rangle, \tag{5}$$

with $\mathbf{K}_b$ equal to

$$(\mathbf{K}_b)_{l,h} = \left(\frac{\partial \mathbf{F}_l}{\partial \mathbf{b}_h} \right)_{\mathbf{b}=\hat{\mathbf{b}}}. \tag{6}$$

In Eqs. (4) and (5) the notation $\langle\rangle$ denotes the expectation value operator. The quantities $(\mathbf{K}_b \varepsilon_b)$ are usually referred to as the error spectra of the FM parameters.

The minimum of the cost function of expression (2) can be found using the Gauss–Newton iterative solution

$$(\hat{\mathbf{x}})_{i+1} = (\hat{\mathbf{x}})_i + (\mathbf{K}^{\mathrm{T}} \mathbf{S}_T^{-1} \mathbf{K})^{-1} (\mathbf{K}^{\mathrm{T}} \mathbf{S}_T^{-1})(\mathbf{y} - \mathbf{F}((\hat{\mathbf{x}})_i, \hat{\mathbf{b}})), \tag{7}$$

where index i numbers the iterations and $\mathbf{K}$ is the Jacobian of the FM, with entries

$$(\mathbf{K})_{l,h} = \left(\frac{\partial \mathbf{F}_l}{\partial \mathbf{x}_h}\right)_{\mathbf{x}=(\hat{\mathbf{x}})_i}. \tag{8}$$

The VCM of the solution is equal to

$$\mathbf{S}_x = (\mathbf{K}^{\mathrm{T}}\mathbf{S}_T^{-1}\mathbf{K})^{-1}. \tag{9}$$

In the OE approach [32] a constrained NLSF is made with the cost function equal to

$$(\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}}, \hat{\mathbf{b}}))^{\mathrm{T}}\mathbf{S}_T^{-1}(\mathbf{y} - \mathbf{F}(\hat{\mathbf{x}}, \hat{\mathbf{b}})) + (\hat{\mathbf{x}} - \mathbf{x}_a)^{\mathrm{T}}\mathbf{S}_a^{-1}(\hat{\mathbf{x}} - \mathbf{x}_a), \tag{10}$$

where $\mathbf{x}_a$ is the a priori estimate of $\mathbf{x}$ with VCM equal to $\mathbf{S}_a$. Accordingly, the iterative Gauss–Newton solution is equal to

$$(\hat{\mathbf{x}})_{i+1} = (\hat{\mathbf{x}})_i + (\mathbf{K}^{\mathrm{T}}\mathbf{S}_T^{-1}\mathbf{K} + \mathbf{S}_a^{-1})^{-1}(\mathbf{K}^{\mathrm{T}}\mathbf{S}_T^{-1}(\mathbf{y} - \mathbf{F}((\hat{\mathbf{x}})_i, \hat{\mathbf{b}})) + \mathbf{S}_a^{-1}(\mathbf{x}_a - (\hat{\mathbf{x}})_i)), \tag{11}$$

and the VCM of the OE solution is equal to

$$\mathbf{S}_x = (\mathbf{K}^{\mathrm{T}}\mathbf{S}_T^{-1}\mathbf{K} + \mathbf{S}_a^{-1})^{-1}. \tag{12}$$

The above equations are common to most inversion problems, however, significant differences are possible in the implementation of the code as discussed in the following section.

5. Wide-band retrieval accounting for FM errors

The main choice faced in the development of the code was between the micro-window approach, with the associated channel selection, and the wide-band approach, with the associated use of the FM VCM.

When redundant information exists on the target quantities, as in the case of most FTS measurements, a micro-window selection can be made in order to minimize the FM errors. In this case the FM errors can be neglected in the retrieval process and one can assume that

$$\mathbf{S}_T = \mathbf{S}_y \tag{13}$$

where $\mathbf{S}_T$ is represented by a block-diagonal matrix with useful saving in both memory occupation and number of operations.

However, in this case the VCM (calculated in the case of the un-constrained solution of Eq. (7))

$$\mathbf{S}_x = (\mathbf{K}^{\mathrm{T}}\mathbf{S}_y^{-1}\mathbf{K})^{-1} \tag{14}$$

accounts only for the measurement noise. The FM errors must be calculated a posteriori and the total error is equal to

$$(\mathbf{S}_x)_{\mathrm{TOT}} = (\mathbf{K}^{\mathrm{T}}\mathbf{S}_y^{-1}\mathbf{K})^{-1} + \sum_b \mathbf{G}[(\mathbf{K}_b\varepsilon_b)(\mathbf{K}_b\varepsilon_b)^{\mathrm{T}}]\mathbf{G}^{\mathrm{T}}, \tag{15}$$

where

$$\mathbf{G} = (\mathbf{K}^{\mathrm{T}}\mathbf{S}_y^{-1}\mathbf{K})^{-1}\mathbf{K}^{\mathrm{T}}\mathbf{S}_y^{-1} \tag{16}$$

is the gain matrix of the retrieval.

As we have already stated, in the case of MARSCHALS a small number of spectral channels is measured and, as an alternative to the micro-window approach, the option can be considered of performing a wide-band retrieval in which all the measured spectral channels are simultaneously considered for a multi-target analysis. The major advantage is to avoid a channel selection which for multi-target retrievals is complicated by the fact that the same channel has different merits for the different targets. A second advantage is the full exploitation of the information content of all the channels, part of which would be disregarded by the channel selection. However, this additional information can be usefully exploited only if the FM errors, which can no longer be considered negligible, are correctly accounted for in the $\mathbf{S}_T$ matrix. FM errors of different channels at different limb-angles are often correlated, and in this case, a full $\mathbf{S}_T$ matrix must be considered, with consequent increased demands of computer memory and computing time.

MARC is designed to perform a wide-band retrieval with rigorous calculation of all the FM errors. FM errors can be caused by the interfering species that are not retrieved, by errors in the instrumental parameters, by approximations in the atmospheric modeling and by spectroscopic errors.

An example of the advantages of this approach is given in Fig. 2 where the retrieval error of the simulated inversion of the MARSCHALS targets is reported. The atmospheric initial guess profiles and the a priori errors of the retrieved targets used for this simulation are reported in Fig. 3. In Fig. 2 the solid line shows the total error (square root of diagonal elements of $\mathbf{S}_x$) of the MARC approach calculated by using Eq. (9), with $\mathbf{S}_T$ given by Eq. (3). This result is compared with both the retrieval error due to the measurement noise calculated by using Eq. (14) (dotted line) and the total error calculated a posteriori by using Eq. (15) (dashed line). The a posteriori calculation of the FM errors leads to a much larger total error than obtained with the MARC approach. This result is not surprising because, in Eq. (9), the FM errors are considered in the retrieval process and a reduced weight is given to spectral channels that are affected by large errors. Instead, a weight that depends only on the measurement error is considered in the case of Eq. (15).

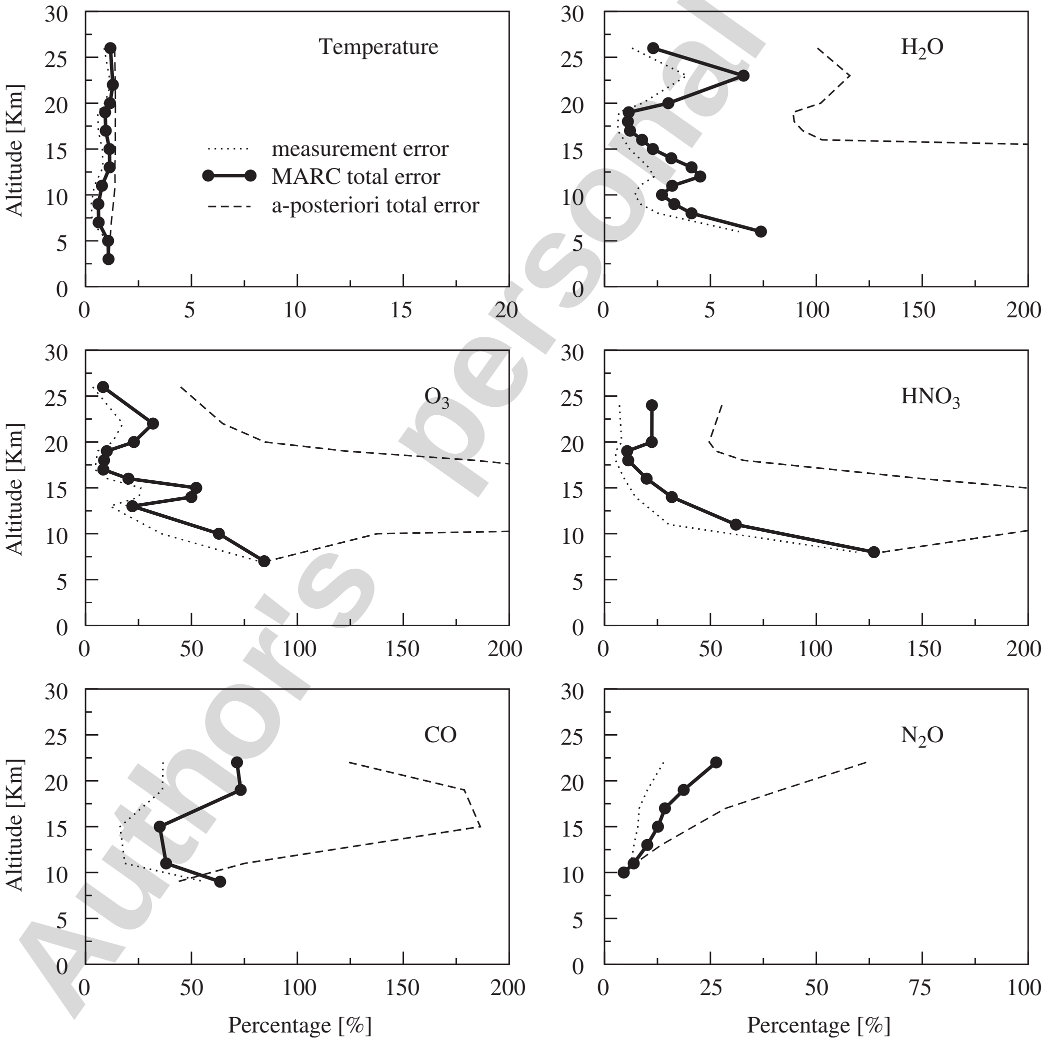

Fig. 2. Error budget of the retrieval targets of MARSCHALS for two different retrieval approaches. The continuous lines with dots show the total error obtained using the MARC approach, in which the retrieval is made with a VCM of the residuals that includes both the measurement noise and the FM errors. The dots indicate the target-dependent retrieval grids used in the test. These errors are compared with the measurement noise retrieval errors (shown by the dotted lines) and with the total error of a retrieval in which the VCM of the residuals only includes the measurement error (dashed lines). In the latter case, the FM errors are added a posteriori to the measurement-noise retrieval errors.

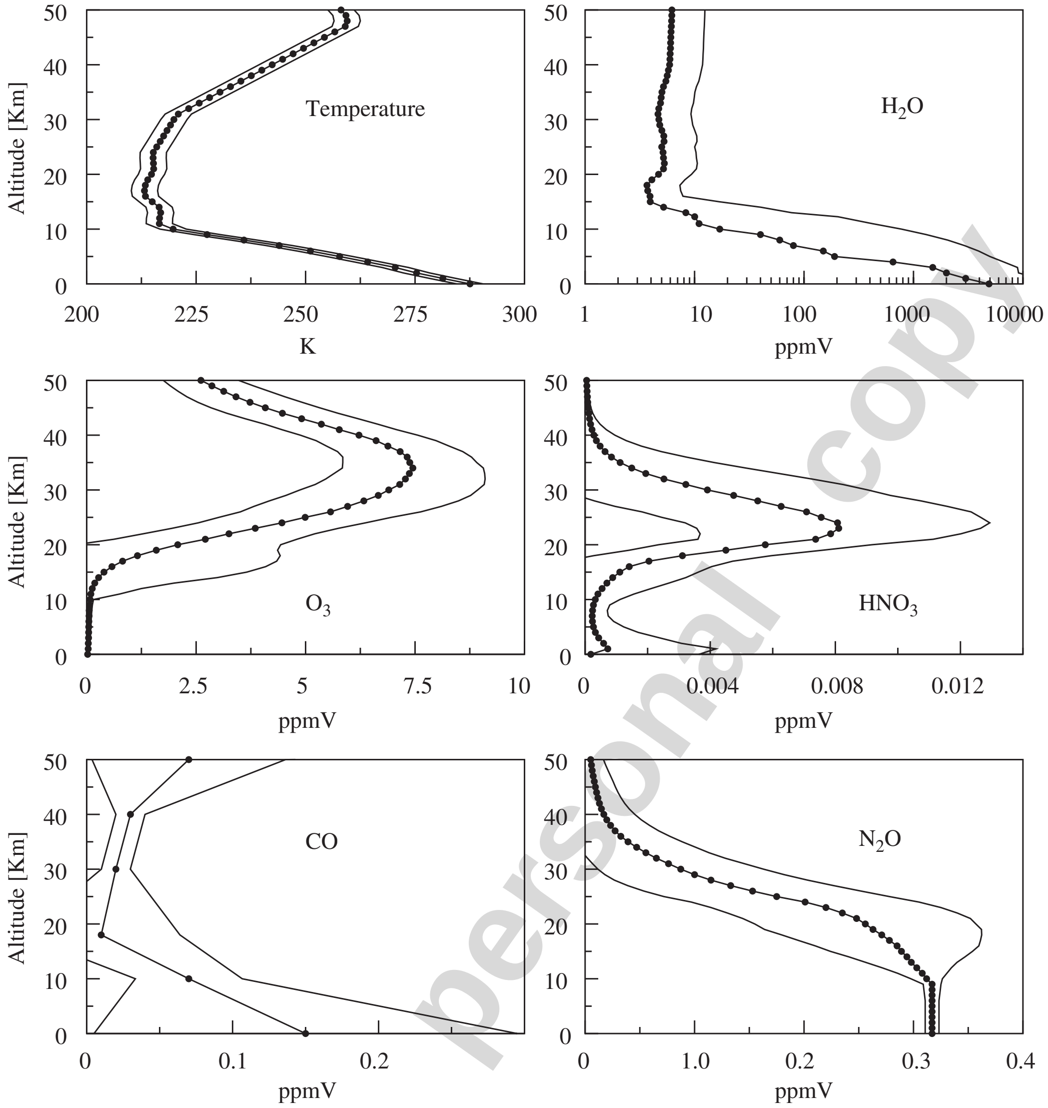

Fig. 3. Atmospheric scenario considered in the simulations of Figs. 2, 4, and 5. Each panel shows the a priori estimates (solid lines with dots) and errors (solid lines) assumed for the atmospheric targets. Mid-latitude climatological conditions are used for the profiles. A typical meteorological uncertainty is assumed for temperature while a climatological uncertainty is assumed for the atmospheric species.

In this comparison, the full MARSCHALS spectral range is analyzed in both cases. This does not correspond to a realistic retrieval made with the micro-window approach, in which a selection of the spectral channels is made. With a channel selection, the total error of Eq. (15) (dashed line of Fig. 2) can be reduced by searching for a compromise between the reduction of the FM error component (second term of Eq. (15)) and the increase of the measurement error components (first term of Eq. (15) and dotted line of Fig. 2). However, the value obtained with this optimization process can never be smaller than the error of the solid line for which all the measured data are optimally exploited.

It is important to underline that the MARC intent and capability of using the full VCM of the FM errors is significantly different from those of existing similar approaches such as the one by Livesey et al. [25] for the analysis of MLS measurements and the one by von Clarmann et al. [33]. These authors consider only the FM errors of the interfering species to calculate $\mathbf{S}_{\mathrm{FM}}$ from Eq. (5), with the objective of limiting the effect of interferences in the retrieval. In the case of MARC all the FM errors are considered in Eq. (5) with the objective of not performing a micro-window selection in a wide-band and multi-target retrieval (see next section).

A disadvantage of the wide-band approach is the increased demand of computing due to the inversion of a full $\mathbf{S}_T$ matrix. Since the number of observations is usually larger than the number of unknowns, this is the largest matrix that has to be inverted in the retrieval. However, with modern computers and fast inversion algorithms, this problem is not a limitation for MARSCHALS that has a relatively small number of observations.

Beside avoiding the difficult process of micro-window selection and reducing the total retrieval error, a further advantage of the wide-band approach is the direct calculation of the total retrieval error that takes into account both the measurement and the FM errors. In practice, with the wide-band approach, the channel selection and the total error calculation are performed directly by the retrieval in an optimal way, and pre- and post-processing calculations are not required.

6. Multi-target retrieval

The choice of wide-band retrieval leads almost directly to the choice of multi-target retrieval. Wide-band retrieval implies that channels are analyzed that contain contributions of several targets. In the presence of mutual interference between the target quantities the best solution is to retrieve all the targets simultaneously.

Furthermore, the recurrent contribution of the instrumental parameters to several spectral channels, when simultaneously considered in a wide-band retrieval, makes the retrieval of these quantities also conceivable. In the case of MARC, a multi-target retrieval is performed in which the retrieved quantities are selected by the user from a wide variety of candidates. Among the possible retrieved quantities there are the scientific targets of MARSCHALS measurements (VMR profiles of the minor atmospheric constituents), complementary atmospheric parameters which must be modeled for a correct description of the observed spectrum (such as temperature, horizontal gradients and continuum absorption) and instrumental parameters.

It may be desirable to obtain for the retrieved profiles of the different targets a different trade-off between vertical resolution and precision. With MARC it is, therefore, possible to choose a target-dependent retrieval grid, the only constraint being that these grids must be a sub-set of points taken from a common fine altitude grid.

The retrieved quantities are individually discussed in the following sub-sections.

6.1. Volume mixing ratio and gradient retrieval

The VMR profiles of H_2O, O_3, HNO_3, N_2O and CO are the scientific targets of the MARSCHALS instrument. However, MARC handles in the same way all the species considered in the FM calculation. In principle, also species that have a very small spectroscopic contribution can be considered as targets of the retrieval.

Usually the vertical profiles of the target species are retrieved ignoring possible horizontal gradients of the VMR. However, since one of the objectives of MARSCHALS is the study of the UTLS region where significant horizontal variability can occur, an horizontal gradient for each species can also be modeled and retrieved as already done in previous codes (see for instance [34]). The horizontal gradient of the VMR is centered at a user-defined geographical location where, by definition, the VMR is not modified by the gradient.

In practice, the VMR profiles and their horizontal gradients are strongly correlated and the retrieved quantities are poorly determined when a conservative a priori information (see next section on OE) based on climatological data is used. Useful results are obtained, however, when the profile is fitted in the presence of a known gradient and when the gradient is fitted in the presence of a known profile. This option will make possible interesting studies with real data in the case of either data merging with other measurements (e.g. the localized measurement of a radiosonde) or orthogonal observations of the same atmospheric air mass obtained with a square path of the aircraft trajectory.

6.2. Temperature retrieval

In the sub-millimeter spectral region, the Planck function of the atmospheric emission is in the Rayleigh–Jeans region and the measurements depend weakly on temperature. The a priori knowledge of temperature obtained from meteorological sources is usually sufficient for the analysis of sub-millimeter measurements. In the case of MARSCHALS, temperature is not a scientific target. However, some information on temperature can be retrieved by exploiting a feature of molecular oxygen which reflects the temperature dependence of the Planck function, and some rotational transitions of vibrationally excited ozone which have a line strength that strongly depends on temperature.

6.3. Continuum absorption retrieval

Continuum absorption due to water vapor, oxygen and nitrogen is considered in the FM of MARC, but further un-accounted absorption effects may occur in a real atmosphere due to an inaccuracy of the adopted models and to the presence of clouds. Therefore, in MARC it is possible to retrieve a further frequency-independent and altitude-dependent absorption coefficient, which is called *fitted continuum*.

The retrieval of the fitted continuum can be used to validate our continuum models in the case of clear sky observations, and to act as a cloud-detection tool once the models have been validated.

The bands of MARSCHALS are rather narrow, and preliminary simulations have shown that the modeling of a continuum that is frequency dependent (in terms of either a slope or a second-order curvature) is not necessary. Therefore, a single constant value is retrieved for each band and each altitude. However, the three bands provide three independent values of the continuum, and these can be used to asses its frequency dependency in a real atmosphere.

6.4. Pointing bias retrieval

Among instrumental parameters, elevation pointing is often critical in the case of limb-sounding measurements. For MARSCHALS, the main source of elevation pointing error is a bias in the attitude of the platform, while it is expected that the relative position of the limb-angles within the limb sequence will be known with good accuracy.

On the basis of these considerations, the capability was built in MARC to fit a constant pointing angle bias for the whole limb sequence and for the three spectral bands. The implicit assumption of perfect band co-registration can be verified with real measurements by performing tests with the individual fit of each of the three bands. If the pointing bias is found to be band-dependent, a pointing correction can be made using a band-dependent offset in the instantaneous field of view (IFOV).

6.5. Instrumental offset and gain retrieval

Other important instrumental parameters are those concerning the radiometric calibration. MARC can be used to retrieve both the offset and the gain of each band. The gain is a channel and limb-angle independent multiplication factor which may deviate from the expected value of unity. The offset is a channel and limb-angle independent additive term which may deviate from the expected null value.

7. The retrieval vector and the use of optimal estimation

The atmospheric species that contribute to the MARSCHALS bands can be retrieved with an accuracy that is target and altitude dependent. Therefore, it is necessary to choose which species are to be included in the retrieval vector and, for each species, which is the optimal altitude range for the retrieval. On the other hand, there are several FM parameters that are an unnecessary constraint if assumed to be known, but that lead to an ill-conditioned retrieval when included among the targets. In general, it is not possible to identify a sharp separation between assumed parameters and retrieved unknowns. This choice is made less critical by the use of the OE technique.

When OE is used, an assumed parameter $\hat{\mathbf{b}}_k$ which contributes to the FM error of Eq. (5) with the VCM

$$\mathbf{S}_{\mathrm{FM}_k} = \langle (\mathbf{K}_{b_k} \varepsilon_{b_k})(\mathbf{K}_{b_k} \varepsilon_{b_k})^{\mathrm{T}} \rangle \tag{17}$$

can easily be transformed into a retrieved unknowns $\mathbf{x}_l$ that has a priori estimate $\mathbf{x}_{a_l} = \hat{\mathbf{b}}_k$ with a VCM

$$\mathbf{S}_{a_l} = \langle \varepsilon_{b_k} \varepsilon_{b_k}^{\mathrm{T}} \rangle. \tag{18}$$

Conversely, the same relationships can be used to transform a retrieved unknown into an assumed parameter.

When OE is used, the retrieval error is given by Eq. (12) which combines the measurement error of Eq. (9) with the VCM of the a priori information. For test purposes, it is useful to compare the retrieval error (which we call *constrained* error because it depends on the a priori information) with the error we would get without using the a priori information (which we call *un-constrained*). For a given retrieved unknown, the ratio between the constrained and un-constrained errors is a quantifier for the impact of the a priori information on the retrieved value and can be used for the choice of the most appropriate retrieval grid. If this ratio is close to one, the retrieved value is mainly determined by the measurements; if the ratio is close to zero, little was added by the measurements to the a priori knowledge and it may be desirable to use a coarser retrieval grid.

Fig. 4 shows the results of a test retrieval for clear sky synthetic measurements in which temperature, O_3, H_2O, HNO_3, N_2O and CO (MARSCHALS targets) were simultaneously fitted together with the pointing bias, atmospheric continuum, offset and gain. The spectra were simulated assuming the atmospheric scenario shown in Fig. 3. The retrievals were performed using OE with the a priori errors shown in Fig. 3 and correlations equal to zero. For each target, the upper frames of Fig. 4 show the differences, between the

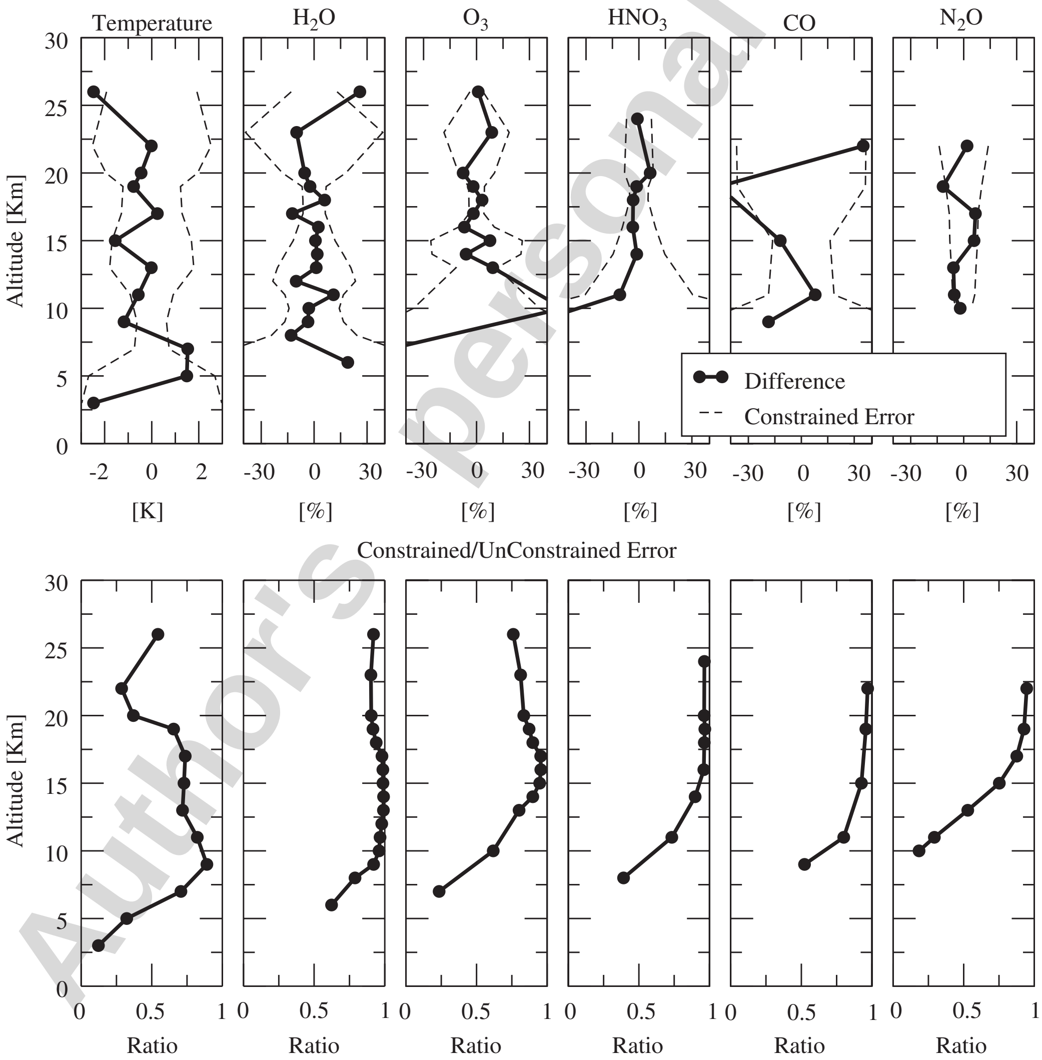

Fig. 4. Results of a test retrieval of MARSCHALS atmospheric targets for clear sky measurements. The marks indicate the target-dependent retrieval grids used in the test. For each target, the upper frame shows the differences between the retrieved profile and the reference profile used for both the generation of synthetic observation and the a priori information. This difference is compared with the constrained retrieval error (dashed lines). The lower frames show the ratio between the constrained error and the un-constrained error. This ratio is a measurement of which the main source of information is between the measurement and the a priori data (see text).

retrieved profile and the reference profile used for the generation of synthetic observations. This difference is compared with the envelope defined by the two dashed lines, which identify the constrained retrieval error and determine the expected accuracy of the retrieval. The lower frames of Fig. 4 show the ratio between the constrained error and the un-constrained error. The retrieved points are identified by a dot, showing that a different retrieval grid has been adopted for the different targets. The retrieval grids are selected with the objective of obtaining a significant contribution of the measurements in the retrieval of the scientific targets. We can see that the ratio is close to one for water vapor between 10 and 17 km, for ozone around 16 km, and for HNO_3 and CO between 15 and 20 km.

For the other targets (pointing bias, offset, gain and atmospheric continuum) an initial guess equal to the actual values, and a very large a priori error were used so that the retrieval was practically un-constrained. These instrumental quantities can be accurately retrieved. The pointing bias was retrieved with an error of 0.008°; the offset was retrieved with an error of 0.15 K and the gain with an error of 0.3%. The atmospheric continuum was retrieved equal to zero, but with a large error. This large error is a sign of possible retrieval instabilities. Indeed, when the initial guess of the atmospheric targets differ too much from the real value, the retrieval may encounter some difficulty in reaching convergence. The problem is overcome when either the initial guess of the atmospheric quantities is close to their real value (as in the test of Fig. 4) or the initial guess of the atmospheric continuum has a smaller a priori error.

von Clarmann et al. [33] have demonstrated that in a single-target retrieval that converges at the first iteration the inclusion of the errors of the interfering species in the VCM of the residuals leads to results that are equivalent to those of an OE multi-target retrieval. A retrieval test has been made in which only the scientific targets of MARSCHALS were retrieved while the temperature, the instrumental parameters, and the atmospheric continuum were considered as assumed parameters that contribute to the FM errors. The retrieved values of the scientific targets are very similar to those of Fig. 4 providing a practical verification of the von Clarmann et al. [33] method also for a multi-target retrieval and proving that the method in some cases is also valid for a retrieval that requires more than one iteration. This implies that in an operational retrieval of MARSCHALS measurements the quantities that are not scientific target can be more efficiently handled as assumed parameters. The capability of fitting these quantities remains, however, an important features that can be used to improve our understanding of the instrumental parameters.

Retrieval tests have also been made in which a single-target retrieval was performed for each of the scientific targets with all the other quantities considered as assumed parameters. The results are shown in Fig. 5 where a comparison is made with the results of the test of Fig. 4. Significant differences are observed, suggesting that the interferences among the target quantities is stronger than that with the instrumental parameters. The strong interference makes significant the non-linear effects and the properties demonstrated by von Clarmann which are valid for a single iteration are no longer verified. The non-linearities of the problem are better handled by the multi-target retrieval. The results of these tests are in agreement with the conclusions reached by Livesey et al. [35] who account for the interference of the temperature error in the VCM of the retrieval, but adopt at the same time a two-step retrieval in order to make sure that a single iteration is sufficient in the second step.

8. The approach to measurements in cloudy scenes

MARSCHALS aims to study the UTLS region and is expected to provide measurements with information also in the presence of clouds [28]. Clouds can cause both absorption and scattering, which contribute to the radiative transfer as a loss and as a further source function (SF). The relative amplitude of the absorption and scattering effects depends on the size of the cloud particles, on their density and on their phase (either solid or liquid). A separate software module called MSSF (*Mie scattering source function*) has been developed in order to take into account the cloud effect. This module calculates the source function of clouds in the millimeter wave region using the Mie theory [36] for a spherical geometry of the Earth's atmosphere with the single scattering approximation. The cloud is modeled as a shell of uniform density, phase (ice and water mixture) and particle size.

MSSF produces the scattering source function in two steps. First, the radiative transfer problem is solved in a spherical geometry for a set of Zenith angles (θ), at each altitude (z) of the atmospheric vertical grid where

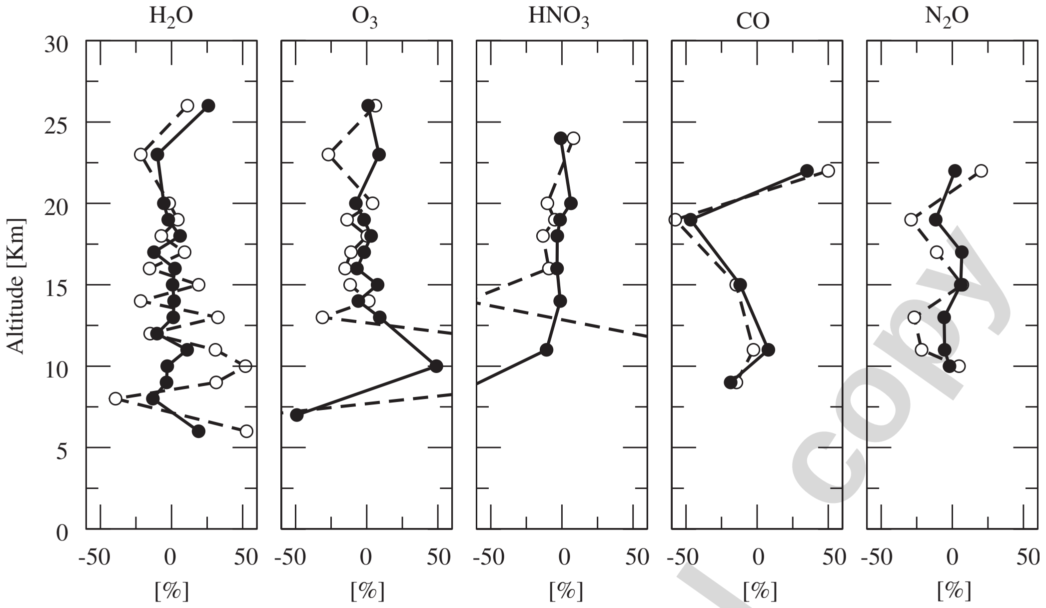

Fig. 5. Difference between the retrieved profile and the reference profile used for the generation of the synthetic observation in the two cases of a single-target retrieval with the errors of the interfering species in the VCM of the FM errors (dashed lines with open circles), and the results of the multi-target retrieval shown in Fig. 4 (continuous lines with close circles).

the cloud is present. In this case, horizontal gradients are disregarded. Accordingly, for symmetry considerations, the calculation is independent of the azimuth angle ϕ. The spectral response of the atmosphere is resolved, and the calculation is performed for each value v of the spectral frequency grid. The result of this first step is the determination of the input field of the radiance $R(z,\theta,v)$ that reaches the scattering particle.

In the second step, the single scattering source function in the horizontal direction is computed by means of angular integration over the full solid angle.

The resulting SF is equal to

$$SF(z,v) = \int_{\theta=0}^{\pi}\int_{\phi=0}^{2\pi} R(z,\theta,v)\sin(\theta)F(\beta(\theta,\phi))\,\mathrm{d}\phi\,\mathrm{d}\theta, \tag{19}$$

where β is the scattering angle, i.e. the angle between the propagation direction of the incident radiation and the horizontal direction, and $F(\beta)$ is the phase function of the scattering process. At the same time, MSSF calculates the absorption and the scattering coefficient introduced by the cloud.

The approximation of a single scattering deserves some considerations. The effect of clouds in the case of limb sounding observation has already been considered by Höpfner [37] and Höpfner and Emde [38] who found that in middle infrared the single scattering approximation can adequately describe for particles of diameters up to about 10 μm the limb measurements with still some useful atmospheric transparency. This result when scaled to the millimeter wave region with the assumption of frequency independent complex refractive index suggests that at long wavelengths the single scattering approximation is valid up to the case of unlikely large particles. However, the absorption coefficient of ice decreases with increasing wavelength and clouds that are opaque in the middle infrared can be transparent in the millimeter wave region so that different cloud densities must be considered in the two frequency ranges and the scaling of middle infrared results cannot be conclusive about the validity of the single scattering approximation. Indeed, comparison with simulations made with millimeter-waves multiple scattering calculations [28] shows that the single scattering approximation leads to errors of about 10% for a cloud with radius particle size of 80 μm and ice mass content of 0.02 $\mathrm{g/m^3}$ which still has some useful transparency at the limb (optical depth of about 1). This ice mass content corresponds to a volume density that is almost three orders of magnitude larger than that of clouds with comparable optical depth in the middle infrared [37]. Therefore, we conclude that, apart from the small error made in the extreme case of clouds with high density and with large particle size, there is a wide range of clouds which are adequately described with a single scattering model, while being opaque in the middle infrared region and transparent in the millimeter wave region.

The calculations of the MSSF module are an option of MARC. When this option is selected, the MARC FM performs the radiative transfer calculations by considering the losses and gains caused by the cloud together with the contributions of the gaseous constituents. The *SF* used in the radiative transfer is computed in the horizontal direction rather than in the actual direction defined by the limb trajectory. This difference is negligible because of the small deviation of the limb-angles from the horizontal direction also considering the smooth shape of the phase function.

The cloud parameters used in the MSSF module can only be used for correction of the cloud effects in the FM, and cannot be selected as target quantities.

9. Sequential estimation

Another important feature of MARC is the possibility of performing a sequential estimation on a set of limb sequences.

We have seen that for some atmospheric constituents the measurements provide very little information when a single limb sequence is analyzed. Furthermore, it may be desirable to determine a more accurate value for some FM quantities that are constant during a flight. In principle, it is possible to retrieve a single target value from a set of limb sequences with the simultaneous fit of all the available observations. However, this implies an increase in memory requirement and on code complexity that we did not want to introduce in MARC, where priority is given to the modeling of FM errors and to the retrieval of a large set of target quantities. The adopted procedure is the sequential estimation with exploitation of the results of the previous retrieval as a priori information for the subsequent one. In this case, a set of limb sequences is analyzed with the desired selection of the targets and, while the target to be averaged is retrieved each time by using the internal a priori information provided by the previous sequence, the other targets are retrieved by using as usual some external a priori information.

Sequential estimation of more than one target is neither a requirement nor a feature of MARC, but the caveats that are necessary in this case deserve some comments. We recall that the correlation between the external a priori information of the different targets is usually equal to zero and it is convenient, as done in MARC, to use a block-diagonal matrix for the representation of matrix $\mathbf{S}_a$ (see Eq. (10)). On the other hand, since simultaneously retrieved quantities are usually correlated, the internal a priori information that must be exchanged in a sequential estimation of several targets requires the use of a full matrix of the a priori errors. In this case the approximation of the block-diagonal matrix can lead to very large errors, even when the correlations are rather small.

10. Conclusion

A new code for the retrieval of atmospheric parameters from limb-sounding spectrally resolved measurements has been presented. The code is called MARC and will be used for the analysis of the observations of the MARSCHALS instrument which will fly on the M-55 stratospheric aircraft. The main objective of this analysis will be the assessment of long-wave measurement capabilities for the study of the UTLS region. Key questions are the feasibility of measuring minor atmospheric constituents in presence of high altitude clouds and, more generally, the measurement quality, in terms of accuracy and spatial resolution.

MARC is a global-fit, multi-target, OE retrieval, in which FM errors are accounted for in the inversion process. It can be used for a wide-band analysis of the observations and its FM can take into account horizontal gradients and cloud contamination. These features make the code optimized for the measurements of MARSCHALS and suitable for several other applications.

The wide-band approach with the calculation of the FM errors as part of the VCM of the residuals provides the full exploitation of the measured information with minimum total error of the retrieved unknowns. Indeed, when the FM errors are used in the retrieval, the total error is smaller than when they are used for a micro-window selection and contribute to the a posteriori calculation of the error budget. In practice, with the wide-band approach the channel selection and the total error calculation are directly performed by the retrieval in an optimum way, and pre- and post-processing calculations are not required.

The OE method makes easy the use of each variable as either an interfering parameter or a retrieved unknown and the multi-target retrieval makes possible the simultaneous handling of many targets.

An example has been shown of a multi-target retrieval in which profiles of six atmospheric targets are retrieved simultaneously with pointing bias, radiometric gain, radiometric offset and profiles of band-dependent atmospheric continuum emission.

The effect of considering some quantities as either retrieved quantities or constant parameters that contribute to the FM errors has also been studied. von Clarmann et al. [33] have demonstrated that for a linear problem the two results coincide. We find that in the case of MARSCHALS the same result is obtained for the scientific targets when instrumental parameters and temperature are considered in either of the two ways. On the other hand, different results are observed for the scientific targets when they are individually analyzed with a single-target retrieval. This difference is explained by the greater role played in this case by non-linear effects.

Using the OE method, MARC can also perform a sequential estimation on a set of limb sequences in order to obtain useful information about those atmospheric constituents which cannot be measured from a single limb sequence.

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