

MODELING AND RETRIEVAL OF ATMOSPHERIC SPECTRA USING ASIMUT

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1. INTRODUCTION

The Earth's atmosphere is continuously changing either on a local or on a global scale. A major challenge for the scientific community is to better understand the involved physico-chemical mechanisms. This requires accurate measurements of trace gases in both the stratosphere and the troposphere. Fourier transform spectroscopy is a powerful technique which is used under various conditions: in ground-based observatories, with balloon-borne instruments, and most recently on board satellites. Inversion of FTIR radiances provides information on the total column abundances, and in some cases on the vertical distributions of several key species.

The ASIMUT software has been developed to exploit the synergy existing between the growing number of different instruments working under different geometries. The main particularities of the software are:

- (i) The possibility to retrieve columns and/or profiles of atmospheric constituents simultaneously from different spectra, which may have been recorded by different instruments or obtained under different geometries. This allows the possibility to perform combined retrieval, e.g., of a ground based measurement and a satellite-based one probing the same air mass, or from spectra recorded by different instruments on the same platform;
- (ii) The analytical derivation of the Jacobians;
- (iii) The use of the Optimal Estimation method (OEM), using diagonal or full covariance matrices;
- (iv) Its portability;
- (v) Its modularity, hence the ease to add future features.

Initially developed for the Earth atmosphere, its applicability can be extended to extraterrestrial atmospheres.

2. SIMULATION MODULE

The ASIMUT program simulates atmospheric transmittances and radiances in cases where scattering can be neglected and under conditions when local thermodynamic equilibrium is verified. The model assumes that all parameters are varying only with altitude. ASIMUT can be used to simulate measurements recorded under various geometries (nadir, off-axis nadir, down and up looking, limb looking) with the instrument located at ground level, inside the atmosphere, or in space.

The equation describing the radiative transfer through the atmosphere can be written as:

$$I(\nu) = I_0(\nu, T_0)e^{-\tau(\nu, 0, s)} + \int_0^s B(\nu, T(s'))\alpha(\nu, s')e^{-\tau(\nu, 0, s')}ds' \quad (1)$$

where I_0 represents the light intensity (often described by the source temperature T_0) at the starting point of the ray path situated at the distance z from the observer, $\alpha(\nu, s)$ is the absorption coefficient, $B(\nu, T)$ is the Planck function, and

$$\tau(\nu, s_1, s_2) = \int_{s_1}^{s_2} \alpha(\nu, s)ds \quad (2)$$

is the optical depth along the path between the points s_1 and s_2 . The integration in Eq. 1 occurs along the ray path, which must be known beforehand.

The simulation of an atmospheric spectrum can be separated into three distinctive components. It first of all requires the determination of the ray path through the atmosphere and the computation of the temperature, pressure and molecular densities along this path. Then, using this information and the line parameters or absorption cross sections relative to each absorbing species, the optical depths corresponding to each layer are determined. Finally, the radiative transfer itself must be solved. The three steps will be described in more details in the following sections.

2.1 Ray tracing

The determination of the radiation path through the atmosphere, i.e. the path followed by the radiation reaching the instrument, requires that the Earth's curvature and refraction be taken into account. The model is based on the ray-tracing program FSCATM [1] improved later when being incorporated into the line-by-line program LBLRTM [2, 3] However some modifications were included in the ASIMUT implementation, essentially dealing with the determination of the index of refraction of air [4].

The user may either choose one of the built-in atmospheres (US Standard Atmospheres) or propose his own volume mixing ratios (vmr) vertical profiles, as well as the vertical profiles for temperature and pressure. The user must further give some of the following parameters to fully describe the ray path: the position of the observer, the looking direction or zenithal angle, the tangent height for limb measurement, and/or the position of the source ending point. The user specifies a layer structure on which the ray-tracing will be performed. Actually, the ray-tracing calculations are carried out on a finer internal grid and the final results, i.e. the effective temperature and pressure for each specified layer as well as the cumulated densities for all involved species, are obtained using the Curtis-Gordon approximations.

2.2 Optical depths

The absorption coefficient $\alpha(\nu, z)$ is in general the sum of several contributions: single absorption lines of different gaseous species, absorption cross sections and continua contributions, and the Rayleigh scattering term.

$$\alpha(\nu, z) = \sum_i n_i \sum_j S_{ij}(T) F(\nu, \nu_{ij}, T, P, p_i) + \sum_c n_c \sigma_c(\nu, T, P, p_c) + \alpha_{\text{Rayleigh}} \quad (3)$$

where P and T are the effective pressure and temperature at altitude z , n_i and p_i the density and the partial pressure of species i at altitude z . The first term of the right hand side of the equation represents the absorption due to single absorption lines. The sum on j represents the sum on all transitions ν_{ij} participating in the absorption of light. The second term represents the broadband absorption of light due to molecules which do not present resolved absorption structures. And the last term is, not *in se* an absorption term, as it corresponds to the Rayleigh scattering, but it is nevertheless taken into account at this point, as it is similar to a diminution of light through the atmosphere.

The intensity $S_{ij}(T)$ exhibits a temperature dependence which can be described as:

$$S_{ij}(T) = S_{ij}(T_0) \frac{Q_i(T_0)}{Q_i(T)} e^{\left(c_2 E_j \left(\frac{1}{T_0} - \frac{1}{T} \right) \right)} \frac{1 - e^{-c_2 \nu_{0j} / T}}{1 - e^{-c_2 \nu_{0j} / T_0}} \quad (4)$$

where c_2 is the second Planck constant hc/k_B (1.4387 cm K), with h the Planck constant, k_B the Boltzmann constant, and c the speed of light, ν_{0j} is the central wavenumber of the j^{th} transition, E_j is the energy of the lower state (cm^{-1}). $S_{ij}(T_0)$, the intensity at the reference temperature T_0 ($\text{cm}^{-1} \text{ molecule}^{-1} \text{ cm}^2$), is obtained from a reference database (HITRAN[5], GEISA[6]), or from a user compiled database. $Q(T)$ and $Q(T_0)$ are the total partition function under local thermodynamic equilibrium at temperature T and T_0 respectively. This function describes the temperature dependence of the line intensity of the transition and is approximated using the formalism of [7]. ASIMUT allows the user to select between different line profiles (Voigt, Rautian, and Galatry), the Voigt profile being the default one.

The grid, on which the profile must be determined, must be fine enough so that the narrowest line is adequately represented. In the upper atmosphere, the Doppler width is the limiting factor for selecting an adequate sampling value. As the Doppler width depends on temperature, it varies with altitude, and the line profile must be sampled with different steps as temperature varies. ASIMUT determines the optimized sampling for each layer. LBL calculations often require the computation of a large number of line shapes over large to very large spectral intervals. The number of points might become prohibitive. However, it can be observed that the line profile changes slower at a distance from the line center than it does near the center. The solution implemented in ASIMUT is the use of a non uniform grid: the step is progressively enlarged as one goes away from the center. At the end, the absorption line shape is interpolated to correspond to the sampling used for the determination of the optical depth (OD). The latter is chosen by considering the step asked for the radiance or transmission simulation and an oversampling factor either provided by the user or chosen such that the OD wavenumber step is a factor 10 smaller than the final radiance step.

Heavy molecules give rise to generally dense absorption spectra, which can not be resolved into single transitions. Similarly, absorption in the visible and the ultraviolet is also characterized by broad unresolved structures. In those cases, one defines the absorption through the cross section, which depends on temperature and pressure. ASIMUT can import cross section data from the HITRAN and GEISA databases, as well as data provided in ASCII by the user. As

those data may be rather scarce in some cases in terms of different temperature and pressure conditions, it was decided to use the data corresponding to the nearest couple (T, P) present in the databases without any interpolation.

For some gases such as N₂, O₂, CO₂, and H₂O, broad absorption features cover wide spectral regions in addition to single absorption lines. These continua of absorption are due to different physical phenomenon: Collision-induced absorption bands resulting from the interaction of molecules pairs such as O₂-O₂, N₂-O₂, N₂-N₂, superposition of line wings in the case of H₂O and CO₂. Different molecular continua are included in ASIMUT. For CO₂ and H₂O continua, the formalism of MT_CKD_1.0 [8] has been implemented.

The Rayleigh optical depth $\alpha_R(\lambda)$ for the layer at the altitude z is given by:

$$\alpha_R(\lambda, z) = N(z) \sigma(\lambda, z) \quad (5)$$

where $N(z)$ is the molecular number density of air in the layer, and $\sigma(\lambda, z)$ is the total Rayleigh scattering cross section per molecule, which may be expressed as:

$$\sigma(\lambda, z) = \frac{24\pi^3}{\lambda^4 N(z)^2} \frac{[n(\lambda, z)^2 - 1]^2}{[n(\lambda, z)^2 + 2]^2} F(\lambda, z) \quad (6)$$

where $n(\lambda, z)$ is the refractive index of air at λ and z ; $F(\lambda, z)$ is the King factor for the depolarization of air at λ and z introduced to take into account the effects due to the anisotropy of air molecules that scatter more radiation at 90° than isotropic ones with the same index of refraction. The F factor is calculated as the weighted sum of the partial contributions due to the molecules of N₂, O₂, Ar, and CO₂, considering that each contribution varies as a function of wavelength. The weights are given by the volume concentration of the atmospheric constituents [9].

2.3 Radiative transfer model

The radiative transfer calculation is performed in each layer following the radiation path. The radiation exiting one layer $I_{k+1}(\nu)$ is obtained from the radiation entering it $I_k(\nu)$, through

$$I_{k+1}(\nu) = I_k(\nu) + B(\nu, T_k) [1 - \tau_k(\nu)] \quad (7)$$

where $B(\nu, T_k)$ is the Planck function calculated at the effective temperature of the layer T_k and τ_k is the transmittance of the layer:

$$\tau_k(\nu) = \prod_i \tau_{ik}(\nu) \quad \text{with} \quad \tau_{ik}(\nu) = e^{-\alpha_{ik}(\nu) n_{ik}} \quad (8)$$

where the product is taken on all absorbing processes i occurring in the layer k ; α_{ik} are the absorption coefficients (cm² molecule⁻¹) and n_{ik} are the cumulated densities in the layer (molecule cm⁻²).

The resulting radiance is then processed to take into account the instrumental line shape (ILS). All conventional ILS of Fourier transform spectrometers (Boxcar, Norton Beer strong,...) are implemented in the program. The field of view effect, as well as the distortion of the ILS by off-axis incident light [10], are included. It is also possible to use a user specified ILS, or to correct the ILS with a modulation efficiency and a phase error [11].

3. RETRIEVAL MODULE

ASIMUT's two main objectives were to use the Optimal Estimation Method (OEM) [12] coupled to the analytical calculation of the Jacobians and to allow simultaneous retrieval from a series of spectra obtained under different geometries, with the possibility to combine spectra recorded by different instruments.

3.1 General description

The general forward radiative transfer equation can be written as:

$$\mathbf{y} = \mathbf{f}(\mathbf{x}, \mathbf{b}) + \varepsilon \quad (9)$$

where \mathbf{y} is the measurement vector (the measured radiance), \mathbf{x} is the state vector (the vertical profiles to be retrieved), \mathbf{b} represents the additional parameters used by the forward model, whose function is \mathbf{f} . The forward function \mathbf{f} describes the complete physics of the measurement, including the description of the instrument. Using the OEM and in the case of a moderately nonlinear problem, the estimate $\hat{\mathbf{x}}$ of the solution of Eq. (9) is found by solving iteratively

$$\hat{\mathbf{x}}_{i+1} = \mathbf{x}_a + (\mathbf{S}_a^{-1} + \mathbf{K}_i^T \mathbf{S}_e^{-1} \mathbf{K}_i)^{-1} \mathbf{K}_i^T \mathbf{S}_e^{-1} [\mathbf{y} - \mathbf{f}(\hat{\mathbf{x}}_i) + \mathbf{K}_i (\hat{\mathbf{x}}_i - \mathbf{x}_a)] \quad (10)$$

where \mathbf{x}_a is the a priori constraint, \mathbf{S}_e the covariance matrix of the measurements, \mathbf{S}_a is the a priori covariance matrix. \mathbf{S}_e is chosen to be diagonal with identical values for all the diagonal elements σ_e^2 , characterizing all the sources of systematic and random errors on the measured radiance. The Jacobian matrix \mathbf{K} is defined by the following expression:

$$\mathbf{K} = \frac{\partial \mathbf{f}}{\partial \mathbf{x}} \quad (11)$$

and will be discussed in great details in the following section. The convergence is attained when

$$d_i^2 = (\mathbf{x}_i - \mathbf{x}_{i+1})^T \mathbf{S}^{-1} (\mathbf{x}_i - \mathbf{x}_{i+1}) \ll n \quad (12)$$

with n the number of parameters to retrieve. The matrix \mathbf{S} is defined as:

$$\mathbf{S} = (\mathbf{S}_a^{-1} + \mathbf{K}_i^T \mathbf{S}_e^{-1} \mathbf{K}_i)^{-1} \quad (13)$$

and represents the total error covariance on the retrieved parameters, which can be decomposed into two contributions: the smoothing error, which accounts for the sensitivity of the measurements to the variable to be retrieved, and the measurement error. Their covariance can be expressed as:

$$\mathbf{S}_{smoothing} = (\mathbf{A} - \mathbf{I}) \mathbf{S}_a (\mathbf{A} - \mathbf{I})^{-1} \quad (14)$$

$$\mathbf{S}_{measur} = \mathbf{G} \mathbf{S}_e \mathbf{G}^T \quad (15)$$

where $\mathbf{G} = \frac{\partial \hat{\mathbf{x}}}{\partial \mathbf{y}}$ is the gain matrix and $\mathbf{A} = \frac{\partial \hat{\mathbf{x}}}{\partial \mathbf{x}} = \mathbf{G} \mathbf{K}$ is the averaging kernel matrix.

3.2 A priori covariance matrices

The a priori covariance matrices (\mathbf{S}_a) should represent the variability of the gas considered. This information is not always available. ASIMUT can manage different type of \mathbf{S}_a matrices: full matrices provided by the user and obtained with the aid of climatologies, diagonal matrices, diagonal matrices with off-diagonal terms added as Gauss or exponential functions

$$S_a(i, j) = \sqrt{S_a(i, i) S_a(j, j)} \exp\left(-\ln(2) \left(\frac{z_i - z_j}{l_c}\right)^2\right) \quad (16)$$

$$S_a(i, j) = \sqrt{S_a(i, i) S_a(j, j)} \exp\left(-\frac{|z_i - z_j|}{l_c}\right)$$

to account for correlations between the concentrations at different altitudes (z_i is the altitude of level i , l_c is the vertical correlation length).

3.3 Determination of the Jacobians

All Jacobians used by the retrieval module are analytically computed during the forward model calculations. The computation is based on the separability of the functions representing on the one hand the radiative forward model $\mathbf{F}_R(\mathbf{x}, \mathbf{b}_R)$ and on the other hand the instrument $\mathbf{I}(\mathbf{b}_i)$:

$$\mathbf{y} = \mathbf{f}(\mathbf{x}, \mathbf{b}) + \varepsilon = \mathbf{I}(\mathbf{b}_i) \otimes \mathbf{F}_R(\mathbf{x}, \mathbf{b}_R) + \varepsilon \quad (17)$$

in which \mathbf{b}_R and \mathbf{b}_i are respectively the model parameters needed for the radiative transfer part and the instrument description respectively. Note that, implicitly, Eq. 17 means that it is not possible to retrieve instrumental parameters, such as modulation or phase error, with ASIMUT.

From Eq. 1, the following derivatives can be deduced:

$$\frac{\partial \mathbf{F}_R}{\partial T_0} = \frac{\partial I_0(T_0)}{\partial T_0} e^{-\tau(v, 0, s)}; \quad \frac{\partial \mathbf{F}_R}{\partial n_{ik}} = \frac{\partial \mathbf{F}_R}{\partial \tau_k} \frac{\partial \tau_k}{\partial n_{ik}} \quad (18)$$

The $\frac{\partial \mathbf{F}_R}{\partial \tau_L}$ derivatives are calculated along the path using relations similar to Eq. 7. The $\frac{\partial \tau_L}{\partial n_{iL}}$ are computed for each species at each altitudes taking into account the several terms (LBL term, cross section, continua). For H₂O, O₂, N₂, and

CO₂, the derivatives of the Rayleigh scattering are included. Moreover, the O₂ continua expressions depends on the N₂ density (and vice-versa), and the corresponding cross-terms are also included.

4. IMPLEMENTATION

The ASIMUT program has been written in the C programming language. The modularity of the program has been one of the driving motives to write our own code. Most options driven by the user's choices may be easily extended. It is for example relatively easy to add a new instrument: the user just has to provide the function to read the spectra and the instrument line function of the instrument to add. Another example is the possibility to add other line shapes, besides the ones already implemented.

As one of the objectives was to be able to run the program either on Windows based PCs or on Unix systems, no graphic possibilities were at first implemented. However, graphics are possible through the opening of a Matlab session. Data and graphic commands are automatically sent to Matlab while the ASIMUT program is running, if the program has detected the presence of the Matlab environment on the machine.

The user controls the program through an user-friendly ASCII file in which he can specify what has to be calculated, saved and plotted (radiance, transmittance, Jacobians, retrieval results), which spectra are analyzed, in which order. It moreover describes the geometries of each spectrum, the spectral intervals, the molecules, cross sections and continua to take into account, and all parameters needed for the treatment of the species (isotope, list of line parameters, file for cross sections, covariance).

5. CASE STUDIES

5.1 Comparison with LBLRTM

The outputs from ASIMUT have been compared to those of LBLRTM under different configurations. Some results relative to the following geometry (observer on ground surface looking at the sun with a solar zenith angle of 30°) are presented here. The vertical profiles describing the atmosphere were chosen identical (US Std atm tropical). Fig. 1 presents the radiances calculated by both programs and their relative differences, and in Fig. 2, are plotted the differences (in %) in effective pressure and temperature, and some species densities. From Fig. 1, it can be observed that the agreement between both programs is very good. The major discrepancies appear when the absorption lines are near saturation or saturated. Some of the discrepancies can be also related to the slight differences existing in the effective densities obtained by both raytracing procedures (see Fig. 2). The latter mainly arise because of the different calculation of the index of refraction and of the dry air density.

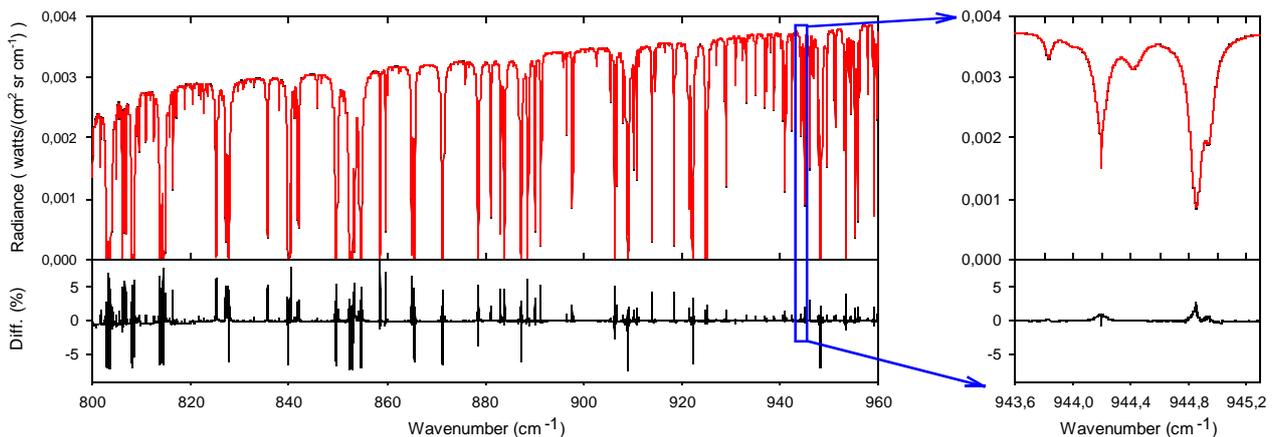


Fig. 1: Comparison of radiances obtained with ASIMUT and LBLRTM

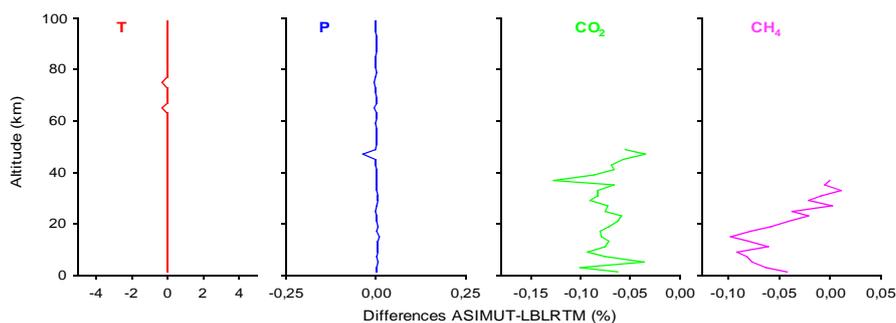


Fig. 2: Comparison of the raytracing results of ASIMUT and LBLRTM: effective temperature and pressure, CO₂ and CH₄ vertical profiles. In the case of LBLRTM, a layer amount is set to 0 if the cumulative amount for that layer and above is less than 0.1 % of the total amount.

5.2 Examples of retrievals

ASIMUT has been tested in several cases studies: retrieval of HCl vertical profile from ground-based occultation spectra recorded at the Ile de la Réunion (using either a Voigt or a Galatry line profile; improved retrieval when a series of spectra obtained at different solar zenith angles are considered simultaneously); retrieval of O₃ from the ACE FTS instrument; retrieval of H₂O, O₃, CH₄, and N₂O from IMG spectra.

6. CONCLUSIONS

The ASIMUT program has proven its capability in modeling atmospheric transmittances and radiances, as well as the possibility to retrieve atmospheric vertical profiles from measured radiances. In the near future, ASIMUT will be coupled to an algorithm allowing the retrieval of aerosol features from nadir thermal radiance spectra. Based on the vertical optical depths obtained with ASIMUT, this algorithm uses an OEM to retrieve both the aerosol optical thickness and the surface temperature, assuming a thin horizontal layer of a mixture of two aerosol types located at an arbitrary altitude.

7. REFERENCES

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