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A fictive gas-method for accurate computations of low-resolution IR gas transmissivities : application to the 4.3 μm CO_2 band

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Résumé. — La méthode proposée de calcul de la transmittivité IR, moyennée sur un intervalle spectral de 25 cm^{-1} , de colonnes gazeuses hétérogènes et anisothermes est applicable aux calculs de télédétection de signature IR et de transferts radiatifs thermiques. Le principe de cette méthode consiste à considérer des gaz fictifs caractérisés chacun par un ensemble de raies d'absorption qui présentent le même comportement en fonction de la température. Les paramètres de ce modèle sont obtenus à partir d'un code de calcul raie par raie ; ils sont tabulés en fonction de la température et d'une largeur de raie adimensionnée. Ce modèle conduit à des résultats beaucoup plus proches de ceux du calcul raie par raie que les modèles statistiques à bandes étroites, et demande des temps de calculs plus réduits que l'approche raie par raie.

Abstract. — The transmissivities of inhomogeneous or nonisothermal gas columns are computed for a 25 cm^{-1} resolution ; the method is suitable for teledetection or coupled radiative and convective transfer applications. The basic idea is to consider fictive gases, characterized by a set of lines of the real absorbing gas, which have a common behaviour with respect to temperature. We use a line-by-line approach to obtain the parameters of our model, which are tabulated as functions of the temperature and of a non dimensional line-width parameter. This model leads to a better agreement with a line-by-line calculation than narrow-band-statistical models and requires smaller computer times than the line-by-line approach.

1. Introduction.

The knowledge of the radiative properties of inhomogeneous and nonisothermal gas mixtures is required in many applications : e.g., temperature measurement through high-resolution methods, tele-detection and IR plume signature, gas-gas radiative transfer calculation... The spectral resolution required varies widely with respect to the application. The Line-By-Line (LBL) approach [1-4] is the most precise, but the most CPU time consuming. Narrow-Band-Statistical models (NBS) [5-10] are rougher, and cheaper ; their parameters can be derived from LBL calculations [4, 11, 12] or directly from experimental results ; reference [12] gives a rather exhaustive bibliography on these models. NBS approaches give accurate results for homogeneous and isothermal gases ; applications to inhomogeneous and nonisothermal media require the approximations of Curtis Godson [13] (CG) or Lindquist Simmons [14] (LS), which are sometimes rather poor.

The original method proposed here allows a description of radiative properties of such media similar to that of NBS models, but without the CG or LS approximations. The objective is to compute,

for any inhomogeneous and nonisothermal column the mean transmissivity $\bar{\tau}_{\nu_1}$ averaged over every adjacent spectral interval of width $\Delta\nu$, centred at ν_1 . Previous publications [11, 12] show that this approach allows a good characterization of radiative properties in all applications considered here. Our model consists in calculating $\bar{\tau}_{\nu_1}$ as the product of transmissivities of a mixture of fictive gases. Each of these gases, labelled c , is defined as having the lines of the real absorbing gas, whose rovibrational lower level energies E'' belong to a given interval $[E_{\text{inf}}^{nc}, E_{\text{sup}}^{nc}]$.

A similar idea can be found in reference [15] to improve NBS models ; nevertheless the authors use the CG approximation, which fails in the applications considered here.

The results of our LBL model have been shown to agree with experiments up to 1 500 K [3, 4]. The remaining problems are due to the relative shortcomings of existing high temperature spectroscopic data (intensities and widths for CO_2 and H_2O , and even line positions for H_2O). An important research effort is still required in that domain. In the present

paper, we intend to validate an approximate model derived from our LBL approach. The validity of this model approximations is determined through a strict comparison with the results of the LBL code, considered as exact. We use the LBL computation in order to generate a set of parameters which lead to a simplified computation of the radiative properties of the gas but which nevertheless are more accurate than the NBS models. In order to describe the CO_2 4.3 μm band (twelve 25 cm^{-1} wide intervals, temperatures from 300 to 1 500 K and total pressures from 0.3 to 3 bars) LBL models require a storage area of 10 Megabytes, our model 10 kilobytes, and NBS models 1 kilobyte. The CPU time required by the method described here for the computation of $\bar{\tau}_{\nu_1}$ is much smaller than that involved by LBL approach.

The method is described, first in the case of an homogeneous and isothermal column in section 2.1, then for the general case in section 2.2. A computational example is given in section 3. Results are compared with those of the LBL and NBS models in section 4.

2. Description of the method.

2.1 CASE OF UNIFORM MEDIA. — Accurate line-positions, which are used in the LBL computation, are unnecessary for low resolution radiative transfer calculations and, consequently, for radiation heat

transfer calculations. A permutation of the spectral positions inside an interval of centre ν_1 and width $\Delta\nu$, over which the blackbody intensity $I_\nu^0(T)$ remains constant, does not affect the computed results. We represent the absorption coefficient K_ν by a monotonous function, noted $k(\nu)$, growing over the interval $\Delta\nu$; this function is derived from the spectrum by successive permutations. Figure 1 illustrates such a transformation. The ordered func-

Fig. 1. — Schematic spectrum : (a) calculated by the LBL approach ; (b) after ordering.

tion $k(\nu)$ may in practice be described by only M values k_m , associated to judiciously chosen spectral positions ν_m . The mean transmissivity $\bar{\tau}_{\nu_1}$ of an homogeneous and isothermal column of length L , i.e.,

$$\bar{\tau}_{\nu_1} = \Delta\nu^{-1} \int_{\nu_1 - \frac{\Delta\nu}{2}}^{\nu_1 + \frac{\Delta\nu}{2}} \exp[-k(\nu) L] d\nu, \quad (1)$$

can then be computed easily using a Gauss-Jacobi or a Gauss-Legendre method.

Introducing the reduced variable :

$$\nu^* = \frac{\nu - \nu_1}{\Delta\nu} + \frac{1}{2}, \quad (2)$$

we can find M couples (A_m, ν_m^*) such that

$$\int_0^1 \exp[-k(\nu^*) L] d\nu^* = \left[\sum_{m=1}^M A_m \exp(-k_m L) \right] + \varepsilon(M) \quad (3)$$

with

$$k_m = k\left(\nu_1 + \Delta\nu \left(\nu_m^* - \frac{1}{2}\right)\right), \quad (4)$$

and where $\varepsilon(M)$ can be made as small as necessary by increasing M . As an example, the CO_2 spectrum which was defined by some 2 500 values of K_ν is represented by typically 7 values of k_m . This method can be generalized to inhomogeneous and nonisothermal columns.

2.2 CASE OF NON UNIFORM MEDIA. — Let us assume that an inhomogeneous and nonisothermal column is discretized into n homogeneous and isothermal radiative elements, quoted i , of length L_i and absorption coefficient K_{ν_i} . The mean transmissivity $\bar{\tau}_{\nu_1}$ of this column is then written :

$$\bar{\tau}_{\nu_1} = \Delta\nu^{-1} \int_{\nu_1 - \frac{\Delta\nu}{2}}^{\nu_1 + \frac{\Delta\nu}{2}} \exp\left(-\sum_{i=1}^n K_{\nu_i} L_i\right) d\nu, \quad (5)$$

where K_{ν_i} results from the contributions of all the lines. An accurate computation of $\bar{\tau}_{\nu_1}$ must take into account the very strong correlations between the line structures of consecutive radiative elements. The spectral line positions are thus invariant with temperature T , total pressure P and absorbing gas molar fraction x , which affect only line-intensities and shapes.

We sort the lines of an absorbing gas into N classes, numbered c , each class containing the lines whose lower rovibrational level energies are in the range $[E_{\text{inf}}'', E_{\text{sup}}'']$. The lines of different classes can be considered to belong to different fictive gases, since the absorption of these lines is not correlated. We associate thus a fictive mean transmissivity $\bar{\tau}_{\nu_1}^c$ to each class and make the same assumption as for absorption by several gases [16]: the mean transmittivity $\bar{\tau}_{\nu_1}$ of an inhomogeneous and nonisothermal column is the product of the mean transmissivities of each class:

$$\bar{\tau}_{\nu_1} = \prod_{c=1}^N \bar{\tau}_{\nu_1}^c. \quad (6)$$

In order to calculate $\bar{\tau}_{\nu_1}^c$, let us consider the absorption coefficient K_{ν_r} of the r^{th} line of class c , in the conditions of a column element:

$$K_{\nu_r}(T, P, x) = x P F_L S_r^0 \frac{1 - \exp\left(-\frac{h\nu_r}{kT}\right)}{1 - \exp\left(-\frac{h\nu_r}{kT_s}\right)} \frac{T_s}{T} \frac{Q(T_s)}{Q(T)} \exp\left[E_r'' \left(\frac{1}{T_s} - \frac{1}{T}\right)\right], \quad (7)$$

where ν_r is the line centre; T , T_s , P and x are the temperature, standard temperature (296 K), total pressure and molar fraction of the absorbant species considered; h and k are the Planck and Boltzmann constants, S_r^0 is the standard intensity of line r , E_r'' its lower level energy and $Q(T)$ the total partition function of the absorbing molecule.

The Lorentz normalized line shape of this line,

$$F_L = \frac{\gamma_r}{\pi [\gamma_r^2 + (\nu - \nu_r)^2]}, \quad (8)$$

is characterized by a half-width (HWHM) in the conditions of an element:

$$\gamma_r(T, P) = \sum_j x_j \gamma_r^j(T, P) = \sum_j x_j \frac{P}{P_s} \left(\frac{T_s}{T}\right)^{\beta_r^j} \gamma_r^{js}, \quad (9)$$

where, P_s is the standard pressure (1 atm), j the collision partner of the absorbing molecule, x_j the corresponding molar fraction, β_r^j a phenomenological parameter and γ_r^{js} the standard half-width. If we define the reduced line-width parameter:

$$G_r^j = \frac{\gamma_r^j(T, P)}{\gamma_r^{js}(T_s, P_s)}, \quad (10)$$

and make the assumption that β_r^j is independent from the line and from the collision partner, we obtain

$$G_r^j \simeq G = \frac{P}{P_s} \left(\frac{T_s}{T}\right)^{\beta}. \quad (11)$$

In most cases, G depends only marginally on the concentration; G can be considered as a nondimensional halfwidth which is usable over a wide range of conditions, even in the case of a model for which the line-width varies from line to line.

We now make the two following assumptions:

i) Substitution of ν_r by ν_1 in (7) leads to negligible error.

ii) $Q(T)/Q(T_s)$ is constant for all lines.

The absorption coefficient due to class c which contains N_c lines then becomes:

$$K_{\nu}^c = \frac{1 - \exp\left(-\frac{h\nu_1}{kT}\right)}{1 - \exp\left(-\frac{h\nu_1}{kT_s}\right)} \frac{T_s}{T} \frac{Q(T_s)}{Q(T)} x P \sum_{r=1}^{N_c} \exp\left[\frac{E_r''}{k} \left(\frac{1}{T_s} - \frac{1}{T}\right)\right] F_L S_r^0. \quad (12)$$

If the spread $E_{\text{sup}}'' - E_{\text{inf}}''$ of a class is small enough, the variations of K_ν^c versus T , at constant G , do not depend on ν . At low resolution, for large optical paths, since line-wings contribute much more than the narrow core of the lines, we can assume, as a first approximation, that the function F_L is proportional to G ; in such conditions, the variations of K_ν^c with G do not depend on ν . In practical calculations, these ideal conditions are not always verified; nevertheless we assume that, for a given class and a given interval, the different ordered parameters k_m , which we obtain as in 2.1 for different column elements characterized by different values of T and G , correspond to the same spectral positions. The spectral correlations for the whole column are hence taken approximately into account by

$$\bar{\tau}_{\nu_1}^c = \sum_{m=1}^M A_m \exp \left[- \sum_{i=1}^n L_i k_{m_i}^c (T_i, G_i, x_i, P_i) \right], \quad (13)$$

where the coefficients A_m associated to the M values ν_m^* are the same as in (3). This last approximation will be justified by the results of section 4.

3. Example of implementation.

The choice of the ν_m^* and the obtention of the corresponding values of A_m derive from the following considerations;

i) A good absolute accuracy on transmissivity is needed in case of weak absorption;

ii) The functions that we have to integrate in (3) are monotonous; their variations are moderated by the exponentiation, so that low degree polynomials are good approximations.

We choose to use seven couples (ν_m^*, A_m) reported in table I, i.e.: $M = 7$. The first five values of ν_m^* belong to the range $[0.0, 0.9]$; we have chosen: $\nu_5^* = 0.9$. The corresponding A_m values are derived from the approximation that $k(\nu)$ is a fourth degree polynomial. The two last values of A_m are such that ν_m^* belongs to the range $[0.9, 1.0]$ and suppose a second degree Legendre polynomial approximation of $k(\nu)$ where A_5 is associated to ν_5^* . We consider five classes of E'' values ($N = 5$), for which E_{inf}'' and E_{sup}'' are given in table II. The

Table I. — Chosen values of ν_m^* and A_m .

m	ν_m^*	A_m
1	0	0.045
2	0.1554058	0.245
3	0.45	0.32
4	0.7445942	0.245
5	0.9	0.06360435
6	0.9436492	0.05008963
7	0.9872983	0.03130602

Table II. — Choice of classes.

Class	E_{inf}''	E_{sup}''
1	0	1 000
2	1 000	2 000
3	2 000	3 000
4	3 000	4 000
5	4 000	∞

relative spread of these classes leads to the following parametrization of k_m^c as a function of T and G :

$$k_m^c = \frac{xP \left[1 - \exp \left(- \frac{h\nu_1}{kT} \right) \right] k_m^{*c}}{T^2 \left[1 - \exp \left(- \frac{T_1}{T} \right) \right] \left[1 - \exp \left(- \frac{T_2}{T} \right) \right] \left[1 - \exp \left(- \frac{T_3}{T} \right) \right]}, \quad (14)$$

with

$$k_m^{*c} = \exp \left[a_1^c + a_2^c \ln(G) + \frac{a_3^c}{T} + \frac{a_4^c}{G} + \frac{a_5^c}{TG} + \frac{a_6^c}{G^2} + \frac{a_7^c}{TG^2} \right]. \quad (15)$$

The equation (14) uses a simplified development of the partition function $Q(T)$ of CO_2 (the chosen

absorbant species), with: $T_1 = 1\,996.0$ K, $T_2 = 960.0$ K and $T_3 = 3\,416.2$ K. The seven coefficients

a_p^c are obtained for the seven values of m , the five classes and twelve spectral intervals of width 25 cm^{-1} centred from 2150 to 2425 cm^{-1} , in the case of CO_2 broadened by N_2 . We have used seventy five high resolution spectra of a column characterized by $x_{\text{CO}_2} PL = 10^{-3} \text{ m} \cdot \text{atm}$; these spectra correspond to three values of T (300, 900 and 1500 K), five values of G (0.1, 0.2, 0.5, 1 and 2) and the five classes defined in table II. In order to prevent discontinuities, the lines used for the calculations are the same for all cases and correspond to the complete set of lines to be considered for the different conditions. The spectral resolution of the computation is optimized for each value of G . In each spectral interval $\Delta\nu$, the spectra are ordered to produce a list of k_m^c values. The a_p^c parameters are then, deduced from (14) and (15) by a least-squares fit on logarithms, limited to values which verify the criterion :

$$k_m^c > 3 \times 10^{-4} \text{ m}^{-1}. \quad (16)$$

This fit introduces a mean relative error of 2.47 % on the values of k_m^c , which verify this criterion ; this is satisfying. In order to demonstrate the quality of the fit, which leads generally to superposed curves, figures 2 and 3 show the worse fit cases ; these curves correspond respectively to couple $m=5$ of the second class and to couple $m=6$ of the third class ; the heavy lines are the k_m^c coefficients, line-by-line computed, and the dashed lines are the values of k_m^c deduced from the a_p^c . The maximum relative error is 20 % (case of Fig. 2).

Figure 4 shows the temperature and pressure ranges for which the radiative properties of the gas have been tabulated. The limits of validity of the Lorentz line-shape are indicated. Nevertheless the

Fig. 2. — Example of k_m^c worse fit case (for $\nu_I = 2350 \text{ cm}^{-1}$, $m=5$ and $c=2$) : — values deduced from LBL approach ; ---- values deduced from equation (15), for five G conditions (0.1, 0.2, 0.5, 1.0, 2.0) and three temperature conditions ((a) 300 K, (b) 900 K, (c) 1500 K).

Fig. 3. — As in figure 2 but with the values : $\nu_I = 2375.0 \text{ cm}^{-1}$, $m=6$, $c=3$.

Fig. 4. — T and G values used in the calculations : ● ; ---- boundary of the range of validity for a Lorentz line shape.

method can be generalized to other line shapes (Doppler, Voigt...).

The colliding partner considered for our calculation is N_2 . However the parameters may be used for other collision partners as long as the variations of the evolutions of the line-width with the rotational number J of the lower level remain acceptable, as it is generally the case.

4. Validation and results.

The FG method is in this section compared with our LBL approach, considered as a reference, and with the NBS model, widely used. We have chosen conditions in which this last model does not lead to good results (large gradients, etc...).

Figures 5 and 6 show a typical use of line-classes in the case of an homogeneous isothermal column of length L ; $\exp(-k^c L)$ is plotted versus ν^* for

Fig. 5. — Examples of ordered functions ; (1) to (5) : ordered expressions of $\exp(-k^c L)$ for classes 1 to 5 ; (6) : ordered transmissivity for the real absorbing gas.

Fig. 6. — As in figure 5.

each class ; the last curve is the ordered transmissivity of the column (which would correspond to the use of only one class). Since the ordering is performed independently for each class, it is clear that the ordered transmissivity of the column is not the point by point product of the functions $\exp(-k^c L)$. Nevertheless the mean transmissivity over $\Delta\nu$, which is the integral of the ordered transmissivity, is computed in our model as the product of the integrals of the $\exp(-k^c L)$ functions ; this fact is the consequence of the hypothesis of no correlation between the lines belonging to different classes.

This last hypothesis, the quality of the calculation through the use of A_m and ν_m^* and the parameter fit resulting of equations (14) and (15) have been checked systematically for all the values of T and G of figure 4. This validation is successful since no significant discrepancy appears except in one case shown in figure 7 ; an intermediate calculation using

Fig. 7. — Transmissivity of an uniform column ($T = 900$ K, $G = 2.0$, $L = 1.0$ m, $x_{\text{CO}_2} = 10^{-3}$, $P = 1$ atm) : — LBL calculation ; ---- our method.

A_m and ν_m^* , but not the parameters a_p^c , agrees with the LBL results ; the error which appears in figure 7 can be attributed to the parameter fit.

We have compared the FG method and a NBS model, which uses an exponential-tailed-inverse distribution intensity law and the Curtis Godson approximation [12]. The band model parameters have been obtained from a fit on LBL results with the procedure described in reference [4] ; the line-width variations are analytically taken into account [12]. We have chosen, for the comparison, the cases of columns made of two or many radiative elements, characterized in table III. These extreme conditions are however representative of a combus-

Table III. — Conditions of figures 8-12.

	Element 1	Element 2	Element 3	Element 4
T (K)	300	1 500	1 500	300
P (atm)	0.303	11	1.1	3.03
$x_{\text{CO}_2} PL$ (mm atm)	0.9	1.5	1.5	0.9
G	0.3	3	0.3	3

Fig. 8. — Transmissivity of a non uniform column (elements 1 and 2 of Tab. III) : — LBL calculation, ---- our model, NBS model with CG approximation.

tion chamber seen through a column of rarefied air, for example. Figures 8-12 show results obtained with LBL, FG and NBS models.

The FG model agrees always with the LBL approach in these figures. The bad performances of the NBS model is due to its poor adaptability to variations of G : in fact, it is the CG approximation

needed by this model which is failing. In order to improve these results a parametrization with T and G of the band model parameters should be used ; this would complicate this approach without achieving the adaptability of ours. In the classical cases, characterized by small G variations, the NBS models give also good results, as shown previously [12].

Fig. 9. — As in figure 7 for elements 1 and 4 of table III.

Fig. 11. — As in figure 7 for elements 1, 2 and 4 of table III.

Fig. 10. — As in figure 7 for elements 2 and 4 of table III.

Fig. 12. — As in figure 7 for elements 1, 2, 3 and 4 of table III but with $x_{\text{CO}_2} PL$ values respectively : 0.45, 0.75, 0.75 and 0.45 mm atm.

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