

## Summary of the CO<sub>2</sub> line mixing package

The package contains the FORTRAN code (largely based on Lamouroux et al., (2015)) to calculate the absorption coefficients for CO<sub>2</sub> molecules perturbed by air, taking into account the line mixing effects. This package is compatible with the recent updates on line-shape parameters of CO<sub>2</sub>, as partially described in Hashemi et al., JQSRT 256, 107283 (2020), which will appear in the HITRAN2020 database edition. Note that the line mixing calculation is being done only for the first ten isotopologues of CO<sub>2</sub> at the moment.

**References:** Lamouroux et al., JQSRT 111, 2321-2331 (2010), Lamouroux et al., JQSRT 151, 88–96 (2015), and references therein. Please cite these references along with the HITRAN paper if you are using the present tools; Gordon et al., JQSRT (2021).

The structure of the package is the following:

### 1) data\_new folder

The folder data\_new includes: i) the spectroscopic data tables separated for each vibrational band and updated using the recent changes on the line-shape parameters of CO<sub>2</sub> in HITRAN2020 database. From HITRAN, a file can be produced per a vibrational band (or sub-bands in the case of asymmetric isotopologues) containing all the corresponding rotational transitions. In the file names ("S10001100001 .dat" for instance), the first number in the file name refers to isotopologue number, followed by global upper and lower quanta. See Lamouroux et al., JQSRT 111, 2321-2331 (2010), for the convention of the file names. Then the files are split by vibrational bands with several additional parameters added.

The structure of input files is more complicated than HITRAN F160 format files because of the large number of parameters. Here are the column widths numbers for each data file:

Col number [1:2], (1) Molecule number as in HITRAN;  
Col number [2:3], (2) Isotopologue number (1=most abundant, 2= second...etc) as in HITRAN;  
Col number [4:15], (3) Wavenumber (cm<sup>-1</sup>) of the line;  
Col number [16:25], (4) Intensity of the line in (cm<sup>-1</sup>/(molecule.cm<sup>-2</sup>)) at 296K;  
Col number [26:35], (5) Einstein-A coefficient;  
Col number [35:40], (6) Air-broadening pressure half-widths (cm<sup>-1</sup>.atm<sup>-1</sup>) at 296K to be used with the Voigt profile;  
Col number [40:45], (7) Speed-dependent Voigt air-broadening half-width (cm<sup>-1</sup>.atm<sup>-1</sup>) at 296K to be used with a speed-dependent Voigt profile;  
Col number [45:49], (8) The speed-dependence of the air-broadening parameters  
Col number [49:54], (9) Self-broadening half-width (cm<sup>-1</sup>.atm<sup>-1</sup>) at 296K using the Voigt profile;  
Col number [54:59], (10) Self-broadening half-widths (cm<sup>-1</sup>.atm<sup>-1</sup>) at 296K using the speed-dependent Voigt profile; This should only have a width of 5  
Col number [59:63], (11) The speed dependence of the self-broadening parameters  
Col number [64:73], (12) The energy of the lower transition level (cm<sup>-1</sup>);  
Col number [73:77], (13) Temperature dependence of the air-broadening half-widths;  
Col number [77:81], (14) Temperature dependence of the self-broadening half-width;  
Col number [81:89], (15) Air pressure shifts of the line transition (cm<sup>-1</sup>.atm<sup>-1</sup>) at 296K;  
Col number [96:144], (16) Transition quantum identifications for the lower and upper state of the transition;  
Col number [151:164], (17) The statistical weight of the upper and lower states;

Col number [164:169], (18) Water broadening half-width ( $\text{cm}^{-1} \cdot \text{atm}^{-1}$ ) at 296K using the Voigt profile;  
Col number [169:174], (19) Water-broadening half-widths ( $\text{cm}^{-1} \cdot \text{atm}^{-1}$ ) at 296K using the speed-dependent Voigt profile;  
Col number [174:179], (20) The water-broadening parameters using the speed-dependent Voigt profile  
Col number [179:183], (21) Temperature dependence of the water broadening half-widths;  
Col number [184:199], (22) Rigid rotor matrix elements;  
Col number [201:215], (23) Lower state relative population.

The folder also contains ii) the list of included bands "bandinfo\_ini.dat" (statistical data for each band) and "Excluded\_bands.dat" (wrong band identification where the calculated absorption coefficient is negative). The file "BandInfo\_ini.dat" gives general information on the bands providing the band identification, its overall integrated intensity, the maximum value of  $J_i$  for each branch (a value of -1 meaning no transitions), and the minimum (Sigmin) and maximum (Sigmax) wavenumbers between which the band contribution will be computed by the main code.

The next is iii) Wfit files which are the off-diagonal relaxation matrix elements files for downward transitions and  $\text{CO}_2$ -air mixtures (for each type of symmetry  $(l_i, l_f)$ ,  $l_f = l_i, l_i + 1$  for 8 temperatures). For a given  $(l_i, l_f)$  vibrational band, the  $W$  off-diagonal elements are then derived as:  $W(T) = W_0(T_0/T)^{-B_0}$ . The first column in these files refers to  $W_0$ , the second is  $B_0$ , the third and fourth one are the maximum relative error (%) in the fitting of  $W(T)$  and the maximum value of  $W(T)$  for each fit, respectively. The last 4 columns are:  $(J_i, J_f)$ , and  $(J_{ic}, J_{fc})$  as coupling between the rotational line  $(J_i, J_f)$  and the line  $(J_{ic}, J_{fc})$ .

## 2) Parameters.inc

The file "parameters.inc" includes some parameters and information on isotopologues used in the main code.

## 3) Main code

The main code for calculating  $\text{CO}_2$ -air absorption coefficient is: "LM\_calc\_CO2.for" and it should be in the same directory with the "parameters.inc", and the "data\_new" folder.

Input parameters which can be changed by the user inside the main code are:

- sgmin, sgmax, dsg [ $\text{cm}^{-1}$ ]: Min and Max wavenumbers and step of the calculation
- sTotMax [ $\text{cm}^{-1}/(\text{molecule} \cdot \text{cm}^{-2})$ ] at 296K]: Total band intensity cut-off
- xCO<sub>2</sub> [no unit]: CO<sub>2</sub> molefraction
- xH<sub>2</sub>O [no unit]: H<sub>2</sub>O molefraction
- Temp [K]: Temperature of the calculation
- Ptot [atm]: Total pressure
- MixFull: Switch to full diagonalization line mixing
- MixSDV: Switch to a Speed-dependent Voigt profile

## 4) The output folder

This empty folder ("output") should be created in the same directory as the main code in order to save the first-order line mixing parameters for each band. For the HITRANonline purpose, we calculated the line mixing at the temperature of 296 K, total pressure of 1 atm, and CO<sub>2</sub> volume mixing ratio of 410 ppm.

Note that the full diagonalization line-mixing is not possible when a speed-dependent Voigt profile is used. Also, for the Q branches and compact regions, the first-order approximation is ineffective, and full line mixing should be included.

In the main code, the user can obtain the absorption coefficient using full line mixing or the first-order line-mixing coefficient by choosing to switch to MixFull or MixSDV. When MixFull is TRUE, then the wavenumber, the absorption coefficient using a Voigt (or speed-dependent Voigt) profile (no line-mixing), the absorption coefficient using a Voigt (or speed-dependent Voigt) profile using the first-order approximation (Y parameters), and the absorption coefficient using a Voigt profile and the full relaxation matrix can be calculated. Following are the output results of the program.

- AbsV ( $\text{cm}^{-1}$ ): Absorption Coefficient neglecting line mixing, using Voigt line shape
- AbsY ( $\text{cm}^{-1}$ ): Absorption Coefficient using the First-order line mixing
- AbsW ( $\text{cm}^{-1}$ ): Absorption Coefficient using Full Diagonalization line mixing