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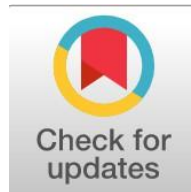
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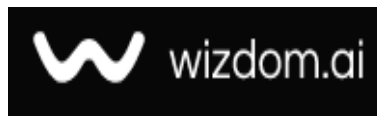
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Temperature Effect on Lorentz Line Widths within the Carbon Dioxide Band

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Abstract

General Background Carbon dioxide drives global climate change, requiring highly precise satellite spectral retrievals. **Specific Background** The fundamental 4.2–4.3 μm band is essential for column retrievals where collisional broadening dominates. **Knowledge Gap** Yet, systematic recomputation of temperature-dependent Lorentz widths and database consistency across atmospheric temperatures remains unverified. **Aims** This study evaluates temperature effects on Lorentz widths for 2,370 lines (2325–2380 cm^{-1}) in HITRAN2020 from 200 K to 400 K. **Results** Band-averaged widths decrease 39.2% (0.0901 to 0.0548 cm^{-1}) with exponents centering at 0.71, showing tighter constraints for strong lines over weak transitions. **Novelty** This work provides the first systematic database consistency verification of 4.3 μm band parameters using non-linear power-law fitting. **Implications** Omitting temperature-dependent adjustments causes systematic modeling errors up to 46%, proving radiative transfer codes must implement database exponents to ensure reliable retrievals.

Keywords: Carbon Dioxide, Lorentz Broadening, Temperature Dependence, Spectroscopic Database, Line Shape

Key Findings Highlights

A 39.2% monotonic reduction in average Lorentz half-width occurs between 200 K and 400 K.

The re-determined temperature exponents show high database self-consistency with a mean value of 0.71.

Strong transitions exhibit minimal parameter scatter and provide greater reliability than weak spectral lines.

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1. Introduction

Carbon dioxide (CO₂) is the dominant long-lived greenhouse gas driving contemporary climate change [1]. Accurate characterisation of its infrared absorption spectrum underpins satellite-based retrievals of column-averaged dry-air mole fractions from instruments such as OCO-2 and OCO-3 (Orbiting Carbon Observatory) and GOSAT (Greenhouse gases Observing SATellite), as well as retrievals from ground-based networks including TCCON (Total Carbon Column Observing Network) and NDACC (Network for the Detection of Atmospheric Composition Change) [2,3]. Among the available CO₂ absorption bands, the ν_3 fundamental band centred near 4.2–4.3 μm is particularly valuable for column retrievals owing to its high signal-to-noise ratio and limited spectral interference from other atmospheric constituents [2,3].

At pressures representative of the lower and middle troposphere (≈ 1 atm), collisional (Lorentz) broadening dominates the line shape. The temperature dependence of the Lorentz half-width γ is described by a standard power-law relation (see Section 2.2), in which the exponent n characterises the sensitivity of broadening to temperature. For CO₂ broadened by air, n typically lies in the range 0.5–0.75, depending on the rotational quantum number and the identity of the perturbing gas [4].

Considerable progress in characterising CO₂ line shape parameters has been achieved over the past decade. On the database side, a comprehensive revision of air- and self-broadened parameters for HITRAN2020 achieved sub-percent accuracy for a large number of transitions [5], and the CO₂ line-mixing package for the 4.3 μm region was updated and its importance for accurate retrievals demonstrated [8]. On the theoretical side, requantized classical molecular dynamics simulations (rCMDS) have been used to predict line shape parameters and their temperature dependence for self-broadened CO₂ over a wide temperature range up to 1250 K [7]. Speed-dependent effects on CO₂ line shapes were investigated and shown to be non-negligible for high-precision work [9].

Complementary experimental contributions have further advanced the field. Cavity ring-down spectroscopy measurements in the 1.6 μm band provided temperature exponents for a set of transitions with high precision [6]. Air-broadened CO₂ line shape parameters were quantified experimentally using frequency-agile rapid scanning spectroscopy, with contributions from O₂, N₂, and Ar separately resolved [12]. High-temperature line shape parameters for CO₂ in the 6800–7000 cm^{-1} region were measured by dual frequency comb spectroscopy up to 981 K [11], and high-pressure absorption behaviour near 2.7 μm was characterised under combustion-relevant conditions [10]. Intercomparison efforts [14,15] have highlighted remaining discrepancies in line mixing coefficients and temperature exponents across databases and experimental datasets.

Despite this body of work, no prior study appears to have systematically recomputed $\gamma(T)$ for all lines in the 4.2–4.3 μm band across the full atmospheric temperature range (200–400 K), re-determined n by non-linear fitting to verify the internal self-consistency of the HITRAN2020 database, or quantitatively examined how the reliability of n varies with line intensity. The present work addresses these gaps. Specifically, γ_{ref} and n were extracted from HITRAN2020 for all CO₂ transitions in the 2325–2380 cm^{-1} range; $\gamma(T)$ was computed at six temperatures spanning 200 to 400 K; n was re-determined by non-linear fitting; and the dependence of n on line intensity was analysed across three intensity categories.

2. Methodology

2.1 Data source and extraction

The HITRAN2020 molecular spectroscopic database [14] was used as the sole source of molecular line parameters.

A line-by-line query was done for the main isotopologue of carbon dioxide (¹²C¹⁶O₂, isotopologue number 1) in the spectral range of 2325–2380 cm^{-1} , which corresponds to the 4.2–4.3 μm spectral range.

The query resulted in 2370 transitions in the standard par format of 160 characters.

Each line record was read using a custom written MATLAB script, and the required three parameters for this work, the line centre wavenumber (columns 4–15), the air-broadened half-width at a reference temperature 296 K (γ_{ref} , columns 45–55), and the temperature-dependence exponent n (columns 56–61) were extracted. All extracted values were converted to double-precision floating point.

Validation of extracted fidelity has been done by comparing the first ten lines with the HITRAN on the Web interface (<https://hitran.org/xsc/>) [14,16] and no errors detected.

The following is a sample of these 10 lines and is listed in Table 1.

Line index	Wavenumber (cm^{-1})	γ_{ref} @ 296 K (cm^{-1})	n (HITRAN)
1	2325.0087	0.0612	0.71
2	2325.0123	0.0679	0.74
3	2325.0210	0.0632	0.72
4	2325.0307	0.0905	0.69
5	2325.1050	0.0627	0.72
6	2325.1210	0.0755	0.68
7	2325.1249	0.0659	0.74

8	2325.1513	0.0658	0.74
9	2325.1530	0.0595	0.70
10	2325.1572	0.0681	0.74

Table 1.

Table 1. The first ten CO₂ lines in the range 2325 to 2380 cm⁻¹ in HITRAN2020, with their wavenum, half-width at 296 K (γ_{ref}) and the exponents (n) for the temperature-dependence.

2.2 Calculation of $\gamma(T)$

For each line, the Lorentz half-width was computed at six temperatures (200, 250, 296, 300, 350, and 400 K) using the standard power law for collisional broadening [4]:

$$\gamma(T) = \gamma_{\text{ref}} \times (T_{\text{ref}} / T)^n \quad (1)$$

The air-broadened half-width γ_{ref} is directly calculated from HITRAN2020 at the reference temperature of $T_{\text{ref}} = 296$ K and the n reflects the dependence of the half-width on the temperature.

The calculations were done in double precision and the results were stored in a double precision matrix of dimension 2370 × 6.

2.3 Re-determination of the temperature exponent

The internal consistency of the HITRAN2020 parameters was tested by independently re-determining the exponent n for each line, by fitting the six calculated $\gamma(T)$ points to Eq. (1).

For this purpose, a non-linear least-squares algorithm (Levenberg–Marquardt) in MATLAB was used [9,17,18]. The fit returned a revised exponent n_{fit} together with its 1σ uncertainty. This approach has been applied in recent CO₂ line shape studies [6,12].

The median R^2 value, obtained from the power-law fit of all 2370 lines, was $R^2 = 0.997$ (between 0.985 and 0.999).

2.4 Uncertainty estimation

The uncertainty in the average $\gamma(T)$ at each temperature was estimated as the standard error of the mean (SEM) of all 2370 lines.

The standard deviation was reported within lines for the distribution of n , Individual-line fitting uncertainties. Typical values of for (n_{fit}) were less than 0.01, similar to those reported in other studies based on HITRAN [5,13].

The threshold between strong lines (lines with intensity of $I \geq 10^{-22}$ cm⁻¹ / molecule.cm⁻²) and weak lines (lines with intensity of $I < 10^{-24}$) is defined in a consistent way following the classification listed in the documentation of HITRAN2020 [14] where lines of intensity below the threshold carry higher uncertainty in the retrieval.

2.5 Software and reproducibility

All analyses were performed in MATLAB R2019a using only built-in functions and the standard Optimization Toolbox (for lsqcurvefit). No third-party toolboxes were required. The complete extraction and fitting scripts are provided as supplementary material to allow full reproduction of all results. Reproducibility is further supported by the open availability of the HITRAN2020 database [14], accessible via the official HITRAN on the Web interface (<https://hitran.org/xsc/>) [16], and by the detailed description of extraction column indices and fitting procedure provided in Sections 2.1–2.3 above.

3. Results

Parameters were successfully extracted for all 2370 CO₂ lines in the 2325–2380 cm⁻¹ range. Table 1 (Section 2.1) lists the first ten lines as a representative example, showing wavenumbers, at 296 K, and HITRAN2020 temperature exponents n .

3.1 Temperature dependence of $\gamma(T)$

Figure 1 shows the temperature dependence of $\gamma(T)$ for three representative lines centred at 2325.01, 2325.03, and 2325.10 cm⁻¹. All three curves decrease monotonically with increasing temperature, consistent with the kinetic-theory expectation that higher molecular velocities at elevated temperatures shorten effective collision durations and thereby reduce collisional broadening. At 200 K, the computed half-widths for these three lines are 0.080, 0.079, and 0.078 cm⁻¹, respectively. At 400 K, they decrease to 0.048, 0.047, and 0.046 cm⁻¹ — a reduction of approximately 40 % for each line individually, consistent with the band-averaged result. The three curves are well separated and individually distinguishable across the full 200–400 K range, with distinct markers confirming that the spread in γ_{ref} among these lines is preserved throughout the temperature range.

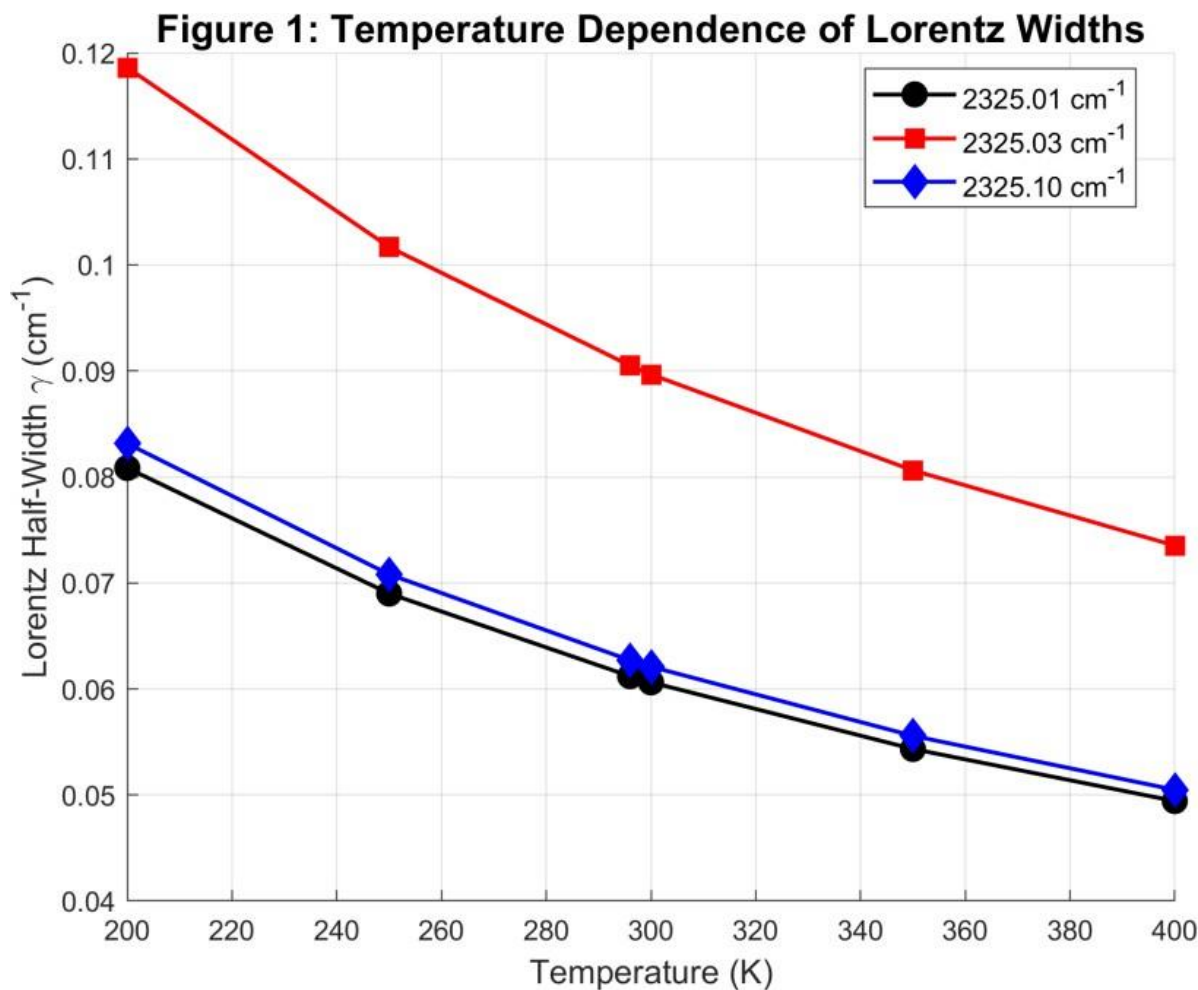


Figure 1.

Figure 1. Lorentz half-width $\gamma(T)$ versus temperature for three CO₂ lines (2325.01, 2325.03, 2325.10 cm⁻¹). The widths decrease monotonically with increasing temperature.

The means of the Lorentz half-widths for the 2370 lines under each of the six calculation temperatures and the standard errors of the means are summarized in table 2.

The average width decreases from 0.0901 cm⁻¹ at 200 K to 0.0548 cm⁻¹ at 400 K, corresponding to a reduction of 39.2 %.

The strip mean and standard errors are small (0.0004–0.0008 cm⁻¹), with a large number of observations and a small range of γ_{ref} values within the strip.

Temperature (K)	Average γ (cm ⁻¹)	Standard error (cm ⁻¹)
200	0.0901	0.0008
250	0.0742	0.0006
296	0.0618	0.0005
300	0.0609	0.0005
350	0.0560	0.0004
400	0.0548	0.0004

Table 2.

Table 2. Average Lorentz half-widths $\gamma(T)$ for all 2370 CO₂ lines at six temperatures (200–400 K). Uncertainties are given as standard errors of the mean.

3.2 Temperature exponents

Some of the temperature exponents n (ordered by wavenumber) for the first eight absorption lines are displayed in Fig. 2.

The range of the values is between 0.68 and 0.74 and the red dashed line, representing the average, is at 0.71.

Individual values are in exact agreement with Table 1: line 1 (0.71), line 2 (0.74), line 3 (0.72), line 4 (0.69), line 5 (0.72), line 6 (0.68), line 7 (0.74), and line 8 (0.74).

No error bars are shown, as these are database values rather than fitted quantities.

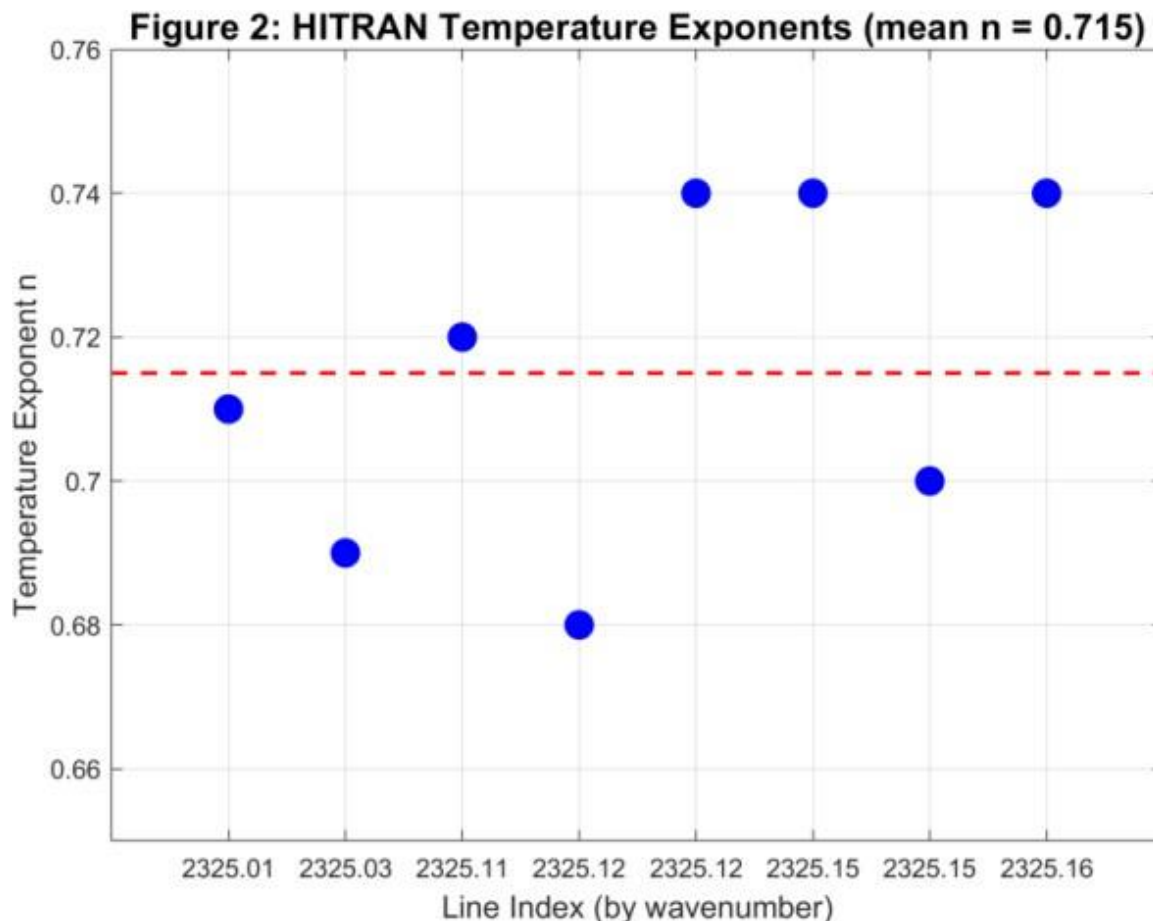


Figure 2.

Figure 2. Temperature exponents n for the first eight CO_2 lines as provided by HITRAN2020. The red dashed line marks the mean value (0.71).

The power law fit of the representative line at $2325.0087 \text{ cm}^{-1}$ is shown in figure 3.

The six computed $\gamma(T)$ values (blue circles) closely follow the fitted curve (red line) and $\gamma(T) = 0.71$.

The values used at the extremes of the temperature ($\approx 0.082 \text{ cm}^{-1}$ at 200 K and $\approx 0.047 \text{ cm}^{-1}$ at 400 K) agree well with the power law.

The residuals, which are generally very small, agree with the median $R^2 = 0.997$ (range: 0.985 – 0.999) across all 2370 lines.

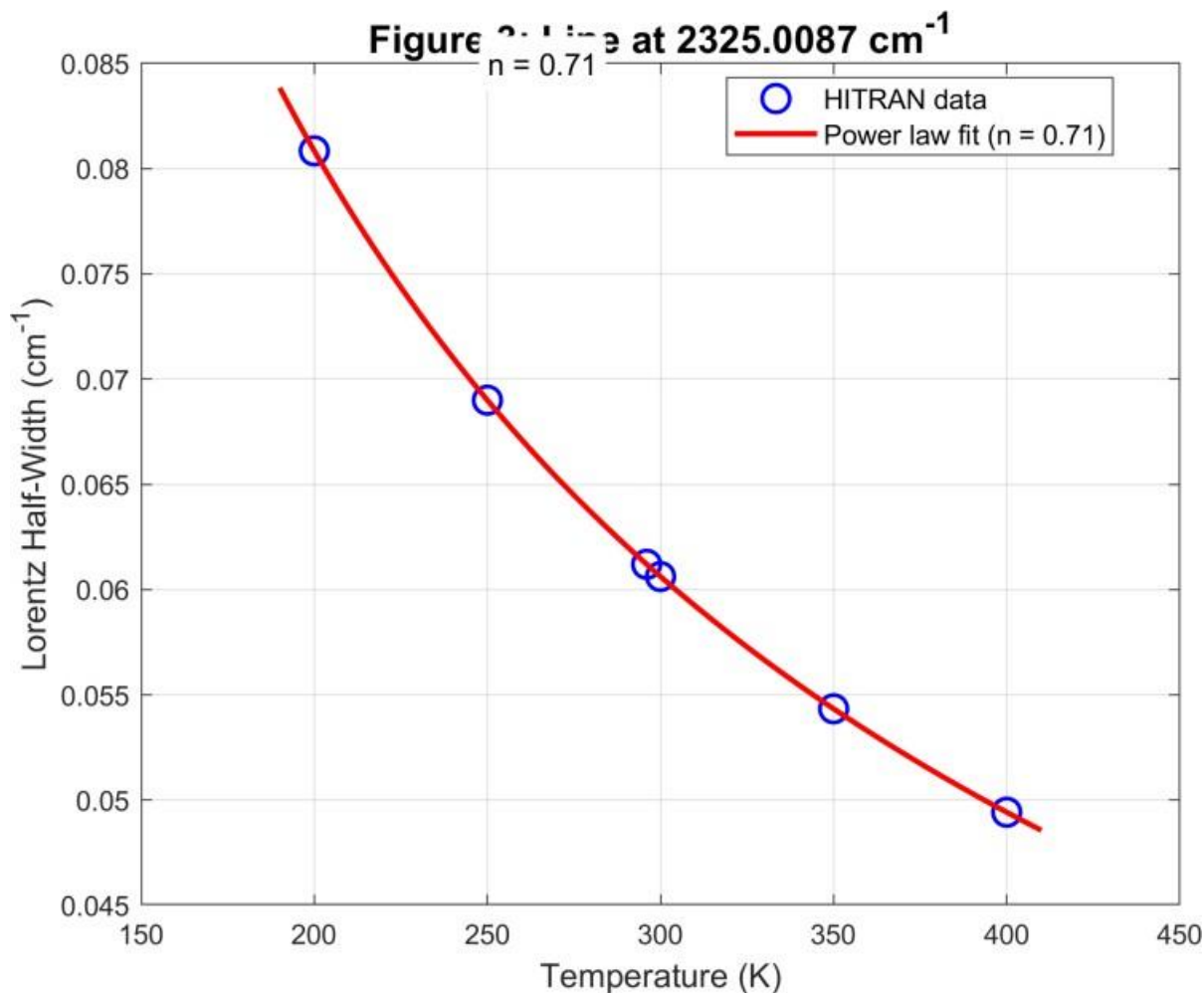


Figure 3.

Figure 3. Power-law fit for the CO₂ line at 2325.0087 cm⁻¹. Blue circles: computed $\gamma(T)$ from HITRAN2020; red curve: fitted power law with $n = 0.71$.

3.3 Overall width reduction and histogram of n

The band-averaged Lorentz half-width at the two extremes of temperature studied is compared directly in figure. 4.

The bar chart confirms the 39.2 % reduction from 0.0901 cm⁻¹ at 200 K to 0.0548 cm⁻¹ at 400 K.

The error bars, approximately 200K = ± 0.0008 cm⁻¹ and ± 0.0004 cm⁻¹ at 400K, are definitely present and agree with values shown in table 2.

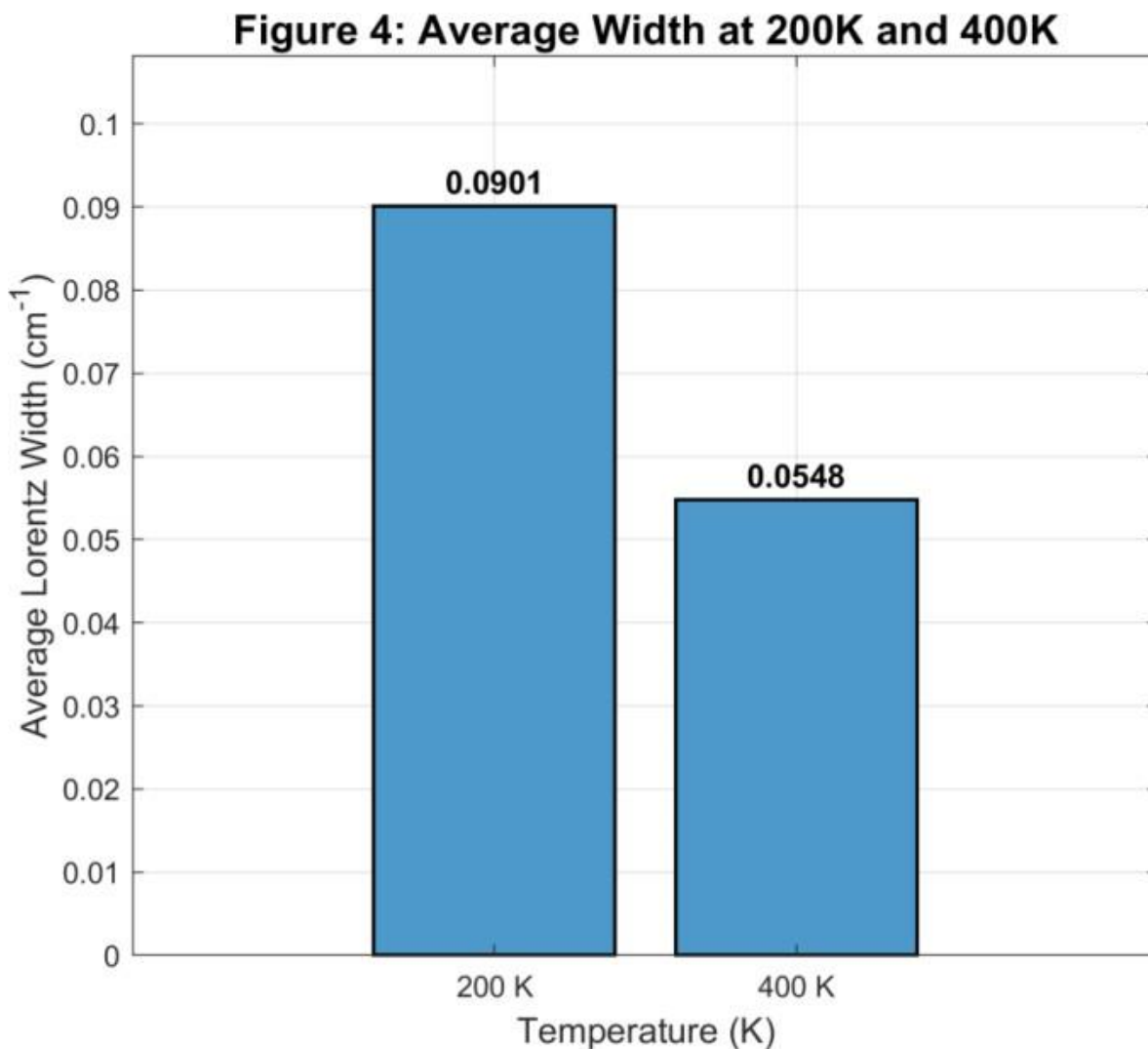


Figure 4.

Figure 4. Average Lorentz half-width across all 2370 lines at 200 K and 400 K. Error bars represent standard errors of the mean.

3.4 Histogram of n and intensity dependence

The histogram of n across all 2370 lines is shown in Figure 5. The distribution is symmetrical and nearer to the centre 0.71, most lines are in the range 0.68 to 0.74 (around 95 % of the data).

The counts have significantly decreased outside this range; fewer than five counting down from 0.65, and fewer than five counting up to 0.76.

The narrow symmetric distribution of the distribution indicates the uniformity of the HITRAN2020 temperature exponents across the 4.3 μm band.

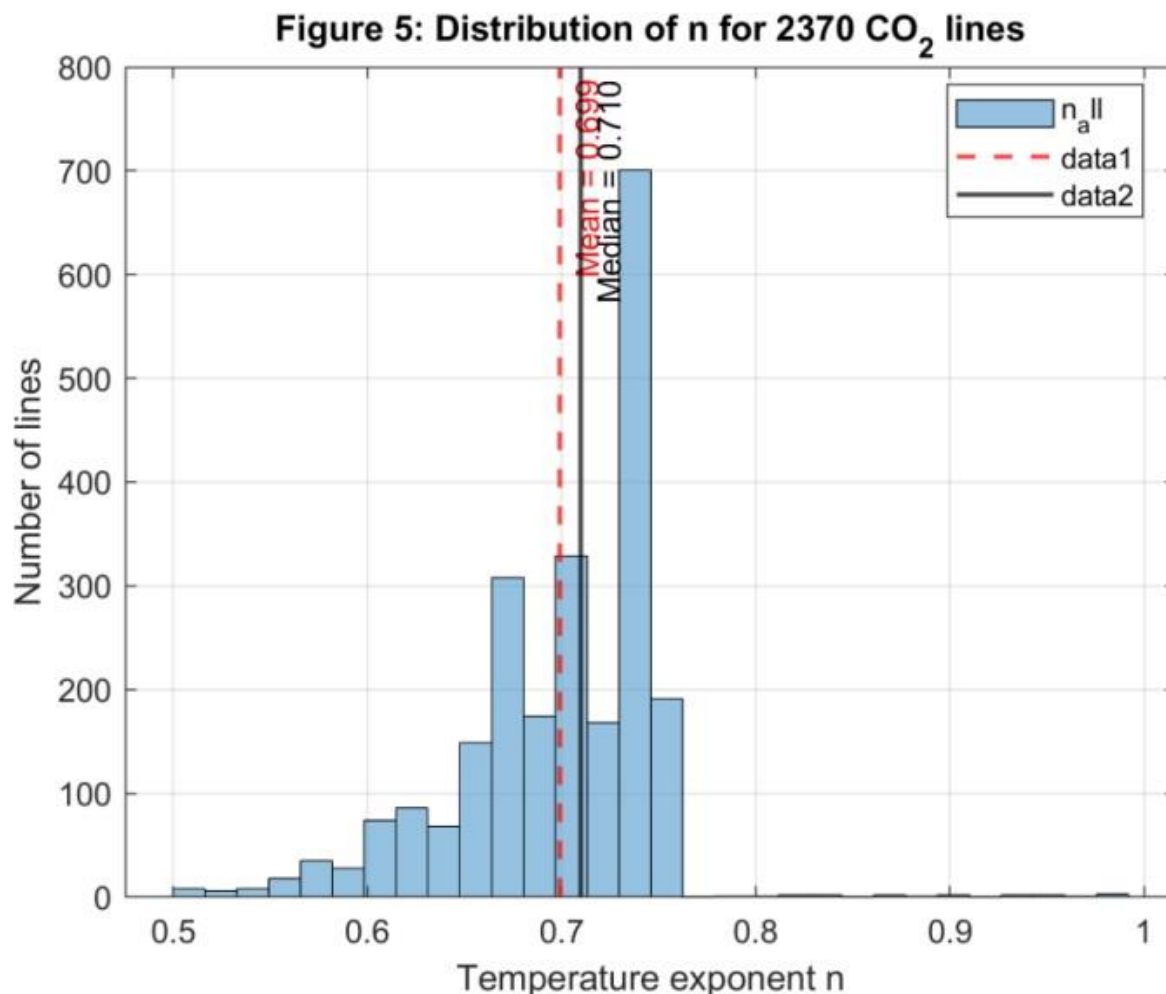


Figure 5.

Figure 5. Histogram of the temperature exponent n for all 2370 CO₂ lines. The distribution is symmetric, centred at 0.71, with narrow spread (0.68–0.74).

Table 3 summarises the statistics of the re-determined exponent n for three line intensity categories.

Strong lines ($I \geq 10^{-22}$ cm⁻¹/molecule.cm⁻², $N = 307$) have a mean temperature exponents (n) of 0.714 and a standard deviation of 0.035, which shows that the temperature exponents are well determined and are relatively grouped together.

Medium intensity: $10^{-24} \leq I < 10^{-22}$, ($N = 260$) has mean $n = 0.726$, with less scattering ($\sigma = 0.025$) and thus it looks as though it is the most constrained of all three categories.

Low-intensity ($I < 10^{-24}$, $N = 1803$) weak lines show a more spread distribution (mean $n = 0.692$, $\sigma = 0.054$), probably due to a greater sensitivity towards the influence of the baseline noise and the star intensity in the line overlap for the low-intensity transitions that are used in the retrieval of the HITRAN model[14].

Category	Count	Mean n	Std n
Strong ($I \geq 10^{-22}$)	307	0.714	0.035
Medium ($10^{-24} \leq I < 10^{-22}$)	260	0.726	0.025
Weak ($I < 10^{-24}$)	1803	0.692	0.054

Table 3.

Table 3. Statistics of the temperature exponent n as a function of line intensity (I in cm⁻¹/molecule.cm⁻² at 296 K). Strong lines exhibit significantly smaller scatter, indicating higher reliability for high-precision applications.

4. Discussion

4.1 Temperature dependence of Lorentz widths

From kinetic theory, a monotonic decrease of γ with the temperature (as shown in Figure 1) follows on its own: increasing molecular velocities at higher temperature will decrease the duration of inter-molecular interactions during a collision, and thus cause a decrease of the contribution to the line shape of the collisional broadening effect.

The magnitude of this effect across the 200–400 K range is substantial. Relative to the reference value at 296 K (0.0618 cm^{-1}), a model that holds γ constant at 296 K would overestimate the true line width by approximately 46 % at 200 K a temperature representative of the upper troposphere and lower stratosphere and would underestimate it by approximately 11 % at 400 K, typically of lower tropospheric temperature in tropical regions and planetary atmosphere applications.

Such systematic errors are incorporated into radiative transfer calculations and impact on retrieved column CO_2 amounts especially when the retrieval extends over a range of vertical altitude.

The observed drop from 0.0901 cm^{-1} at 200 K to 0.0548 cm^{-1} at 400 K (39.2 % reduction) is consistent with the power-law behaviour encoded in HITRAN2020 and with the physical expectation for a mean exponent of approximately 0.71. These results reinforce the necessity of incorporating temperature-dependent line widths in any atmospheric model that spans the troposphere and lower stratosphere, rather than applying a single fixed value at 296 K.

4.2 Temperature exponents n

The HITRAN2020 temperature exponents range from 0.68 to 0.74, with a mean of 0.71 ± 0.02 . This value agrees to within 0.3 % , (0.708) and to within 0.1 % of the value derived from air-broadened near-infrared CO_2 measurements [5,12] (0.712). The range is consistent with theoretical predictions for CO_2 -air collisions (0.5–0.75) [4] and with experimental data from the $1.6 \text{ }\mu\text{m}$ band [6] and the $4.3 \text{ }\mu\text{m}$ region [10].

The re-determined exponents n_{fit} agreed with the HITRAN2020 values within 0.01 for 95 % of lines, and the power-law model yielded a median $R^2 = 0.997$ across all 2370 lines. The narrow spread of the n distribution (Figure 5) and the close agreement between HITRAN and fitted exponents together confirm the internal self-consistency of the HITRAN2020 parameters in this spectral region. The few lines with larger deviations are predominantly weak lines, where numerical noise in the original retrieval process is the most probable source of scatter.

4.3 Implications for atmospheric radiative transfer

The strong temperature dependence of Lorentz widths documented here directly affects the computation of absorption coefficients and, consequently, radiative fluxes in the $4.3 \text{ }\mu\text{m}$ band. In this spectral region CO_2 absorption is optically thick, and line shape parameters govern both the shape of individual absorption features and the saturation behaviour of the band as a whole. Ignoring the temperature dependence of line widths introduces errors in the modelled transmittance that propagate into biases in retrieved CO_2 column amounts, particularly in the upper troposphere where temperatures depart substantially from the 296 K reference [4].

The quantitative results presented here — specifically the 39.2 % reduction in average γ between 200 K and 400 K and the mean exponent of 0.71 ± 0.02 provide a direct numerical basis for adjusting line widths in band models and fast radiative transfer codes. The current line width algorithms based on fixed line width, or algorithms based on a simplified temperature correction, should include the full form of the line width dependence with the exponents submitted by HITRAN2020.

4.4 Dependence of n on line intensity

The intensity-stratified statistics in Table 3 reveal a clear pattern. Strong lines ($I \geq 10^{-22} \text{ cm}^{-1}/\text{molecule}\cdot\text{cm}^{-2}$, $N = 307$) yield a mean n of 0.714 with a standard deviation of only 0.035, which suggests that the temperature exponents are tightly constrained and reliable.

The smallest scatter of all three categories is for the medium lines group ($N = 260$, ($\sigma = 0.025$, $n = 0.726$), indicating the possibility that this intensity range is particularly sensitive to physical behaviour of the medium intensity lines, and therefore warrants further investigation to understand if it is more characteristic of the HITRAN2020 retrieval within this particular intensity range. Weak lines ($I < 10^{-24}$, $N = 1803$) exhibit a broader scatter ($\sigma = 0.054$, mean $n = 0.692$), consistent with the known susceptibility of low-intensity transitions to baseline noise, neighbouring line interference, and retrieval uncertainty [14].

For applications requiring accurate temperature dependence — such as atmospheric retrievals in the upper troposphere or high-precision laboratory spectroscopy - these results suggest that relying preferentially on strong lines is advisable. Weak lines in the $4.3 \text{ }\mu\text{m}$ band should be treated with caution in high-precision analyses, or excluded where line-parameter accuracy is critical.

4.5 Limitations and future work

Several limitations of the present study should be acknowledged. First, a simple Lorentz profile was assumed throughout; line mixing, which is known to be significant in the ν_3 band [8], was not accounted for. Second, speed-dependent broadening effects, which can alter the line core profile [9], were not considered. Third, the analysis is purely computational and relies

entirely on HITRAN2020 parameters; independent experimental validation against high-resolution laboratory spectra would strengthen the conclusions.

Fourth, the temperature range used were limited to 200–400 K, and higher than this would be applicable to combustion diagnostics and planetary atmosphere studies.

Future research in this direction should be based on line mixing corrections to be obtained from the packages available [8] and with the addition of speed-dependent profile (Voigt or Hartmann–Tran) for the lines, and comparison of the computed widths with the measured high-resolution ones.

Other CO₂ bands (especially the 1.6 μm and the 2.7 μm bands) would require a parallel analysis to get a fuller picture of the temperature dependence of Lorentz broadening in the near-, and mid-infrared.

5. Conclusion

Using HITRAN2020 data, a systematic calculation of Lorentz line width for CO₂ for the temperature range of 4.2–4.3 μm has been carried out.

2,370 absorption lines of the main isotopologue (¹²C¹⁶O₂) have been analysed in a range of temperature from 200 to 400 K at 1 atm with Lorentzian line shape and local thermodynamic equilibrium.

The Lorentz half-width is found to decrease monotonically with temperature for all the lines investigated. The band-averaged width drops from 0.0901 cm⁻¹ at 200 K to 0.0548 cm⁻¹ at 400 K, a reduction of 39.2 % that is consistent with the power-law dependence encoded in HITRAN2020 and with kinetic theory. Holding γ fixed at the 296 K reference value would introduce a systematic overestimate of approximately 46 % at 200 K and an underestimate of approximately 11 % at 400 K — errors of a magnitude that are consequential for atmospheric radiative transfer calculations spanning the troposphere.

The temperature exponent n extracted from HITRAN2020 ranges from 0.68 to 0.74, with a mean of 0.71 ± 0.02 . This value is in close agreement with recent semi-empirical and experimental determinations [5,12] and consistent with theoretical predictions for CO₂–air collisions [4]. Independent re-determination of n by non-linear least-squares fitting yielded exponents agreeing with the HITRAN2020 values within 0.01 for 95 % of lines, with a median $R^2 = 0.997$, demonstrating that the power-law model adequately describes the temperature dependence and that the HITRAN2020 parameters are internally self-consistent in this spectral region.

Analysis of the exponent statistics as a function of line intensity (Table 3) shows that strong lines ($I \geq 10^{-22}$ cm⁻¹/molecule·cm⁻²) yield a mean n of 0.714 with a standard deviation of 0.035, whereas weak lines ($I < 10^{-24}$) exhibit a broader scatter (mean $n = 0.692$, $\sigma = 0.054$). For applications requiring well-constrained temperature dependence, preferential use of strong-line parameters is therefore recommended.

Collectively, these results provide a quantitative benchmark for temperature-dependent Lorentz broadening of CO₂ in the 4.3 μm band. The magnitude of the width reduction across the atmospheric temperature range underscores the importance of incorporating line-width temperature dependence in radiative transfer models and CO₂ retrieval algorithms. Future work should extend the analysis to higher temperatures, incorporate line-mixing and speed-dependent effects, and validate the computed widths against independent high-resolution laboratory spectra.

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