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PREDICTION OF TOTAL EMISSIVITY OF CO₂ IN TROPOSPHERE BY SEVERAL METHODS

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ABSTRACT

The effects of CO₂ ppm in troposphere on both spectral and total emissivity are reviewed using several methods. The compared models are by Hottel [2], Bliss [3], Atwater and Ball [4, 6], wide Band Model by Edwards [5], Yamamoto and Sasamori [6,7], and using HITRAN data base [8]. For spectral emissivity, the results by Yamamoto and Sasamori match well with predictions using HITRAN data base. For total emissivity, the deviations between models are rather large and sometimes more than about 0.05. In general, for a given condition, the upper bound of total emissivity is given by Hottel, and lower bound is given by HITRAN. The predictions by Edwards are in between but near to those of Hottel. The CO₂ ppm varied from 300 ppm to 600 ppm, temperature varied from 220K to 300K, and pressure from 0.3 to 1.0atm. As CO₂ ppm increases, the total emissivity increases. For a given CO₂ ppm, the total emissivity increases as the air thickness increases which are also true for both temperature and pressure increase. Around 260K, the total emissivity is less sensitive to increasing temperature than to decreasing temperature. For a given pressure change, the total emissivity values by wide band model changes more than the values by HITRAN. The reverse is true for temperature change.

KEY WORDS

CO₂, Spectral emissivity, Total emissivity, Wide Band Model, HITRAN, Troposphere.

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NOMENCLATURE

E	: Emissive power (W/m^2)
L	: Path length (cm, m)
P	: Pressure (atm)
R	: Specific gas constant (kJ/kg-K)
T	: Temperature (K)
u_c	: Pressure path length product (atm-cm)
X_c	: Density path length product (g/cm^2)

Greek

ε	: Emissivity
γ	: Bliss absorption coefficient ($50\text{cm}^2/\text{g}$)
κ	: Absorption coefficient (cm^{-1} , m^{-1})
λ	: Wavelength (μm , cm)
ρ	: density (kg/m^3 , g/cm^3)
σ	: Stefan Boltzmann constant ($5.67 \times 10^{-8} \text{ W/m}^2\text{-K}^4$)

Subscript

b	: Blackbody
c	: CO_2

INTRODUCTION

Due to the global industrialization, the carbon dioxide (CO_2) concentration keeps increasing. The CO_2 concentration in 2011 was about 380ppm, while it was about 300ppm in 1960. The rate of increase is about 1.5ppm per year [1]. As CO_2 concentration increases, it is predicted that the global warming will be accelerated and it could bring out many disasters. To decrease its level, work should be done to decrease its production and also try to captivate CO_2 . To predict the effects on climate, it is necessary to know the emissivity of CO_2 . Also, to determine the effective sky temperature [3,4,11] the emissivity of CO_2 should be available.

There appear many experimental and theoretical works for total emissivity of CO_2 gas in the literature [2-7,9]. The total emissivity for participating gases including CO_2 was measured and modeled by Hottel [2]. The results are presented in the form of charts, tables, and weighted sum of gray gases model. Edwards [5] measured and modeled the total emissivity of participating gases using wide band model. Smith [9] et al. generate weighted sum of gray gas model for total emissivity of CO_2 . In atmospheric science, the participating gas molecular absorption data base has gone through many modifications, improvements, and enhancements compiled at the recent version of HITRAN [8]. Lallemand et. al. [10] evaluated the total emissivity correlations for $\text{H}_2\text{O-CO}_2\text{-N}_2/\text{air}$ mixtures modeling for high temperature combustors and emphasized the limitations of using one and two weighted sum of gray gases components. However, the results of these works show a large deviation [10].

The purpose of this paper is to predict the spectral and total emissivity of CO₂ gas in the troposphere using various known models and to compare the result. The effects of CO₂ ppm, air layer thickness, temperature, and pressure are studied. Only air and CO₂ are the constituents of the atmosphere and both are assumed to be ideal gases.

MODELS FOR CO₂ EMISSIVITY

The spectral emissivity experimental results and models are diverse and plenty in the atmospheric science literature. In this paper Yamamoto and Sasamori [6,7] and Atwater and Ball [4,6], Bliss [3], and prediction using HITRAN data base [8] are compared. The total emissivity models compared are Hottel [2], wide band model of Edwards [5], and prediction using HITRAN data base [8]. The Hottel and Edwards models are used in combustion engineering whereas the HITRAN is used in the atmospheric sciences.

The total emissivity is defined as:

$$\varepsilon_c = \int_0^\infty (1 - e^{-k_\lambda L}) E_{b\lambda} d\lambda / \sigma T^4 \quad (1)$$

The mole fraction of CO₂ is called CO₂ ppm, and sometimes it is called the volume mixing ratio. The partial pressure of CO₂, P_c , can be expressed by total atmospheric pressure and CO₂ ppm.

$$P_c = P \times (\text{CO}_2 \text{ ppm} \times 10^{-6}) \quad (2)$$

The mixing ratio of CO₂ is the mass of CO₂ per air mass in the atmosphere.

Bliss Model

The total emissivity model used by Bliss [3] is given by Eq. (3). Only 13~17 μm CO₂ absorption band is considered, and the constant 0.185 is the blackbody energy fraction within the band. Without the constant 0.185, it can be regarded as the spectral emissivity of the CO₂ within the band.

$$\varepsilon_c = 0.185(1 - e^{-\gamma X_c}), \quad X_c = \rho_c L = P_c L / R_c T \quad (3)$$

Where the constant $\gamma=50$ [cm²/g], and X_c [g/cm²] is the density and path-length product, ρ_c is the density of CO₂, and L is the thickness of the atmosphere.

Model Used by Atwater and Ball

Atwater and Ball [4] used the 15 μm band CO₂ spectral emissivity model by Shekter [4,6] after changing the coefficient 0.3919 to 0.32 to predict the sky temperature. The spectral emissivity model is:

$$\varepsilon_c = 1 - e^{-0.3919 u_c^{0.4}}, \quad u_c = P_c L (\text{atm} - \text{cm}) \quad (4)$$

Eq. (4) is valid only for the CO₂ 15μm band which spectral wave number region 550-800cm⁻¹ [6]. The total emissivity can be obtained using Eq. (1). For the temperature range between 220K and 300K, the blackbody energy fraction within the band varies from 0.1902 to 0.1837. But the blackbody energy fraction is chosen as the same as that of Bliss [3] for comparison purposes in Ref [12]. The Eq. (5) is a gray approximation.

$$\varepsilon_c = 0.185(1 - e^{-0.3919 u_c^{0.4}}) \quad (5)$$

Yamamoto and Sasamori's Spectral Emissivity Model

The Yamamoto and Sasamori [6,7] made a summary of the experimental and theoretical investigations results for the 15μm [550-800cm⁻¹] band absorption function that is the same as the spectral emissivity of 15μm band. In Figure and Table [6,7], the band absorption function is presented as a function of the density corrected CO₂ path-length u at 1atm total pressure in Eq. (6). For the state at P=1000mb and T=300K, the superscript o is used in Eq. (6).

$$u = \int \rho_c dL / \rho_c^o \quad (6)$$

The results by Yamamoto and Sasamori [6,7] match well with most probable curve from $u=0$ up to $u=10$ [cm at 1atm] but under predicts afterwards. The spectral emissivity results in Ref. [6] are compared in Table 1.

The total emissivity can be evaluated by performing spectral integration as in Eq. (1).

Hottel's Total Emissivity Model

Hottel [2] did experiments to determine CO₂ total emissivity. The results are presented using Tables and Figures in the literature. The lowest temperature is 293K. The results by Bliss [3] are used for comparison purposes in [12].

Wide Band Model by Edwards

The total emissivity can be evaluated using the wide band model by Edwards [5]. For this paper only 15μm CO₂ absorption band being considered whereas all the CO₂ bands such as 15, 10.4, 9.4, 4.3, 2.7, and 2.0μm bands are used in Ref. [12]. The relative error of 15μm band only case relative to whole band case is at most 5.6% for the range of values tested in this paper. The wide band model use 3 parameters such as band width parameter, line width to spacing parameter, and band intensity [5]. After band absorptivity is calculated using these 3 parameters, it is weighted by the blackbody band energy fraction to determine the total emissivity. However, for spectral emissivity values, 80% of the band absorptivity is used according to the suggestion by Edwards [5].

HITRAN Data

HITRAN band data [8] for CO₂ is used to predict spectral emissivity and total

emissivity. The computation procedure used in this paper is as follows: First, for each line data, both the maximum value of absorption coefficient at the line head and line half width are evaluated using the formula in Ref. [8] which considers temperature, pressure, and air broadening effects. Second, it is assumed that each line starts from the line center wave number minus line half width and ends at the line center wave number plus line half width. Third, the line absorption is approximated as the product of absorption coefficient at the line head and line width instead of integrating Lorentz profile [8]. Fourth, to consider line overlap effect, each line's lower and upper limit is checked and new wave number (or length) interval block is generated. In general, total number of the blocks is greater than the total number of lines. At each i^{th} block, if lines overlap, then the following line transmission overlap relations are used to calculate the block transmissivity. The procedure is somewhat like block calculation used by Edwards [5] for wide band model.

$$\tau_{ith} = \tau_o \prod \tau_i \quad (7)$$

For each block, initial block transmissivity is adjusted such that $\tau_o=1$ or 0.95. For small $P_c L$, $\tau_o=1$ is used and on the other hand 0.95 is used for $P_c L$ greater than about 1 atm-cm. Fifth, the spectral emissivity is calculated using Eq. (8)

$$\varepsilon_c = \sum_{ith} (1 - \tau_{ith}) \Delta \lambda_{ith} / \Delta \lambda_c \quad (8)$$

Sixth, the total emissivity is calculated using Eq.(9)

$$\varepsilon_c = \sum_{ith} (1 - \tau_{ith}) \Delta E_{b,ith} / \Delta E_{b,c} \quad (9)$$

The input values used for 15 μm band are, wavelength 13~17 μm [588-769 cm^{-1}], where all 4 isotope component of CO_2 [8] is considered. Only the lines with intensity greater than 10^{-24} [1/atm-cm] (cut-off intensity) are used. There are a total of 3917 lines with this cut-off intensity.

RESULTS

Spectral Emissivity

In Table 1, the spectral emissivity predictions by Yamamoto and Sasamori [6,7], Bliss [3], Atwater and Ball [4,6], wideband model by Edwards [5], HITRAN [8] block calculation results are presented by varying partial pressure path length product, $P_c L$. Two values of block calculation results appear in Table 1, one with initial block transmissivity of 1 and the other with 0.95. The conditions used for the tabulated results are such that total pressure is 1 atm and temperature is 300K.

According to the results in Ref. [6], Yamamoto and Sasamori [6,7] data match well with the most probable curve values up to $u=1$ [cm at 1atm] but afterwards under predict the most probable curve values. As suggested by Edwards [5], the spectral values in Table 1 are 80% of wide band model prediction. Wide band predictions by

Edwards are not accurate for $P_c L$ less than around 300 atm-cm. In general, Bliss [3] and Atwater and Ball [4,6] model do not match well with the predictions by Yamamoto and Sasamori [6,7] and HITRAN [8], except certain range of values of $P_c L$.

HITRAN block calculation results with initial block transmissivity of 1 match well with Yamamoto's results up to $P_c L$ around 0.3 atm-cm. From $P_c L$ around 0.3 up to 100 atm-cm, the initial block transmissivity is adjusted, such as 0.95 in Table 1. But for $P_c L$ over 100 atm-cm, the results by HITRAN are speculated to be more accurate because Yamamoto's results under predict as shown in Ref. [6]. If so, the wide band results by Edwards seem to under predict in this region. Thus, more investigation is needed for $P_c L$ over 100 atm-cm.

Total Emissivity

In Fig. 1, the total emissivity predictions by several models are presented to compare the accuracy. The horizontal axis is density-path length product, X_c , and the vertical axis is the total emissivity of CO_2 . The total emissivity increases as X_c increases except for the results by Bliss [3] for X_c over 0.1. The conditions used are CO_2 300ppm, total pressure of 1 atm, and temperature at 293K. The gray gas model in Eq. (5) [12] derived from the model used by Atwater and Ball [4,6] matches well with the results given by Hottel [2]. In general, the upper bound of total emissivity is given by Hottel, and the lower bound is by HITRAN [8]. The predictions by Edwards are in between but near to those of Hottel. The deviations among results by Hottel and Edwards [5] are about -15% to 10% for the range of values X_c in Fig. 1. The deviation between wide band model and HITRAN prediction increases as X_c increases. The total emissivity is higher at the initial block transmissivity of 0.95 than that at 1.0

At $P=1$ atm, $T=293$ K, CO_2 300ppm, the thickness of the air layer is $L = 36.4, 182.2, 1822$ m at $X_c = 0.002, 0.01, 0.1$ g/cm², respectively. In terms of $P_c L$, it is 1.09, 5.47, 54.7 atm-cm, respectively.

In Fig. 2, the effects of CO_2 ppm on total emissivity are presented at 300ppm, 380ppm, and 600ppm CO_2 . The total pressure is 1atm at the temperature of 293K. The horizontal axis for air layer thickness and the vertical axis for the CO_2 total emissivity. At constant air layer thickness, as the CO_2 ppm increases the total emissivity increases. Also, at constant CO_2 ppm, as thickness of air layer increases, the total emissivity increases. The upper bound is the gray gas in Eq. (5), the lower bound is HITRAN [8], and in the middle is the wide band model by Edwards [5]. All three models show approximately the same increments of total emissivity as CO_2 ppm increase except the gray gas model results beyond 3km.

In Fig. 3, the effects of air temperature on CO_2 total emissivity are studied at the total pressure of 1atm and the CO_2 concentration of 380ppm. The temperature varied from 220K to 300K with increments of 20K. The horizontal axis is for air layer thickness, and the vertical axis is for the total emissivity of CO_2 . At a constant air layer thickness, as temperature increases, the total emissivity increases except for the gray gas model in Eq. (5). Also, the emissivity differences between wide band model and HITRAN increase as air layer thickens. The sensitivity of total emissivity

with temperature increases as air layer thickness increases for both wide band model and for HITRAN, except air layer thickness less than around 10m. However, at a constant air layer thickness, the sensitivity of total emissivity with temperature decreases as temperature increases from 220K to 300K for both wide band model and for HITRAN. Especially for wide band model, the increment of emissivity due to temperature change from 220K to 240K is more than the increment due to temperature increase from 240K to 300K. Also, the result lines for 260K, 280K, and 300K almost collapsed into a single line in Fig. 3 for wide band model.

The results for 300ppm and 600ppm cases show similar trends as in Fig. 3 for 380ppm case, and thus the results are not presented in this paper. At the air layer thickness 1, 100, 10000m, the corresponding $P_c L$ is 0.00679, 0.679, 60.79, respectively, at total pressure of 1atm, temperature of 300K, and at 380ppm.

In Fig. 4, the effects of total pressure on total emissivity of CO₂ are presented at the temperature of 300K and 380ppm CO₂. The total pressure values are 0.3, 0.6, 0.8, and 1atm. The horizontal axis is for the air layer thickness and the vertical axis is for the total emissivity of CO₂. The following trend is as common for all three models, such that at a constant air layer thickness, as pressure increases, the total emissivity increases for all three models. The emissivity listed in higher order at a fixed air layer thickness is gray gas, wide band model, HITRAN with initial block transmissivity of 0.95, and HITRAN with initial block transmissivity of 1.0. The total emissivity values by two HITRAN cases are both less sensitive to pressure change. On the other hand, the reverse is true for the values by wide band model and by the gray gas model in Eq. (5). For both wide band model and gray gas model, the increment of emissivity due to pressure increase from 0.3 to 0.6atm is more than the increment due to pressure increase from 0.6 to 1.0atm.

At the temperature of 220K, the results show similar trends as that observed at 300K. Thus, the result presentation is omitted.

CONCLUSIONS

The effects of CO₂ ppm, air thickness, temperature, and total pressure on both spectral and total emissivity for CO₂ in troposphere are studied using several methods. For the range of values tested, the results show the following trends: Among spectral emissivity models, results by the Yamamoto and Sasamori [6,7] matches well with HITRAN [8] block calculation results. Among total emissivity models, the results by Hottel [2] and gray gas model [12] match well. The total emissivity differences among models are rather large and sometimes more than about 0.05. For a given condition, the total emissivity in highest order is given by Hottel, by Edwards [5], by HITRAN with block transmissivity of 0.95, and by HITRAN with block transmissivity of 1.0. The sensitivity of total emissivity with temperature change is positive and is higher for HITRAN results than for wide band model. The reverse is true for the sensitivity of total emissivity with pressure change. If total emissivity is sensitive to either pressure or temperature, the sensitivity is nonlinear to these variables.

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Table 1. Comparison of spectral emissivity.

PcL (atm-cm)	Yamamoto [6,7]	HITRAN [8]	HITRAN [8](0.95)	Edwards [5]	Bliss [3]	Atwater [4,6]
0.01	0.0080	0.0077	0.0573	0.1902	0.0009	0.0602
0.03	0.0170	0.0188	0.0679	0.3009	0.0027	0.0919
0.1	0.0430	0.0421	0.0900	0.3512	0.0089	0.1445
0.3	0.0880	0.0733	0.1196	0.3732	0.0264	0.2150
1	0.1660	0.1264	0.1701	0.4096	0.0855	0.3242
3	0.2660	0.2097	0.2492	0.5462	0.2351	0.4557
10	0.3870	0.3286	0.3622	0.6169	0.5908	0.6263
30	0.4860	0.4388	0.4669	0.6547	0.9315	0.7830
100	0.5840	0.5685	0.5901	0.6827	0.9999	0.9156
300	0.6650	0.7025	0.7173	0.7019	1.0000	0.9784
1000	0.7510	0.8353	0.8435	0.7184	1.0000	0.9980

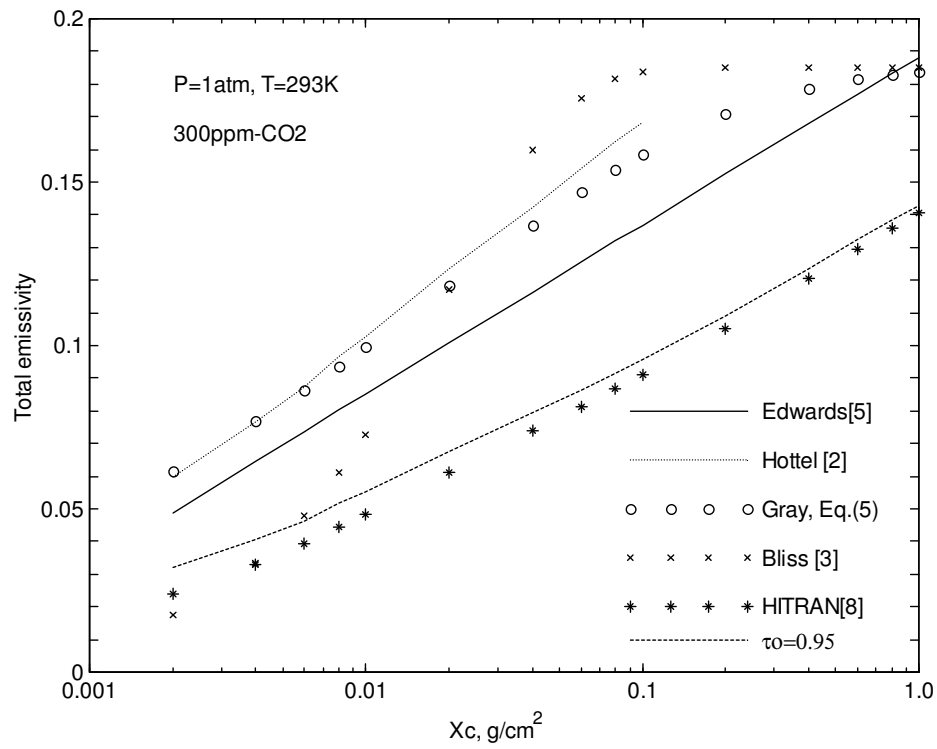


Fig. 1. Effects of $X_c = \rho_c L$ on total emissivity of CO₂

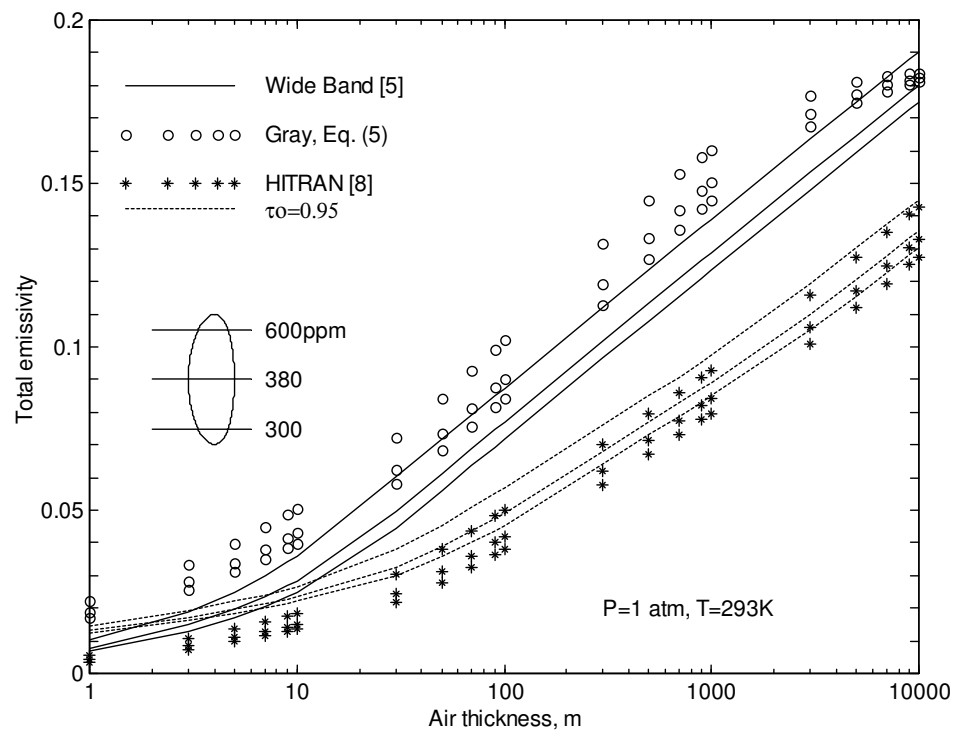


Fig. 2. Effects of CO₂ ppm on total emissivity of CO₂

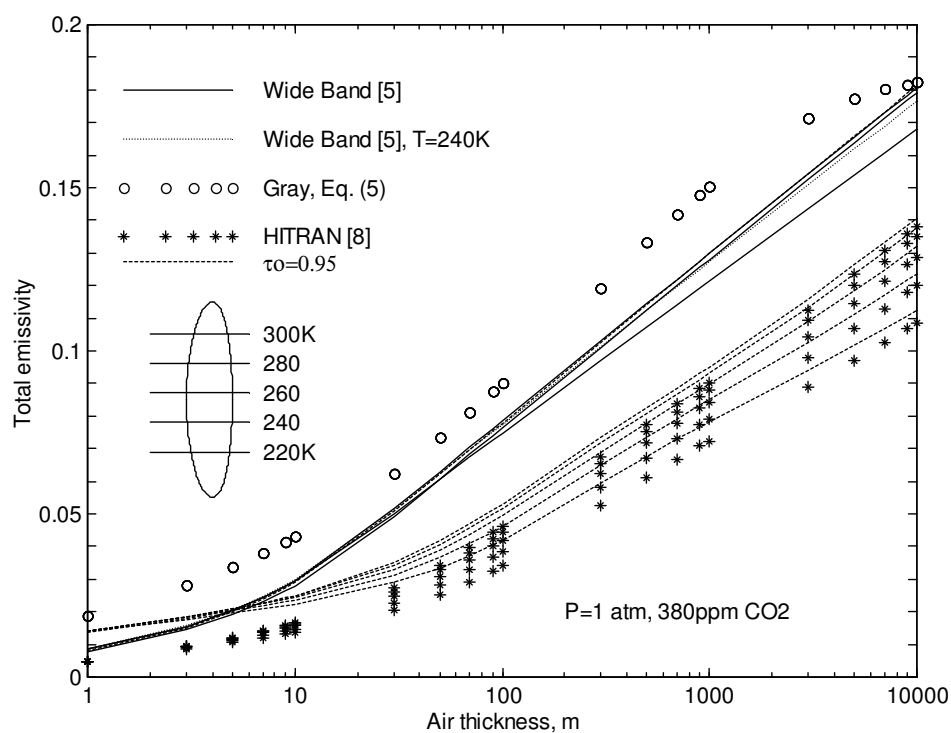


Fig. 3. Effects of temperature on total emissivity of CO₂

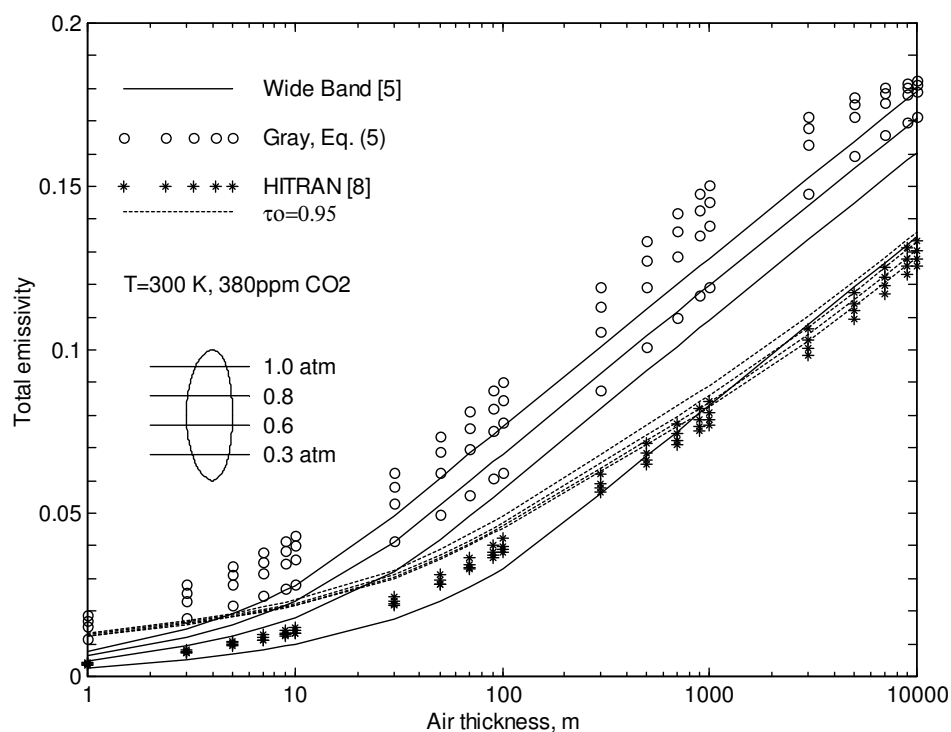


Fig. 4. Effects of pressure on total emissivity of CO₂