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ABSTRACT

This report summarizes the tools and methods used to parameterize the radiative absorption by CH₄ and N₂O within the context of the infrared radiative transfer model of Chou and Suarez (1994), currently being used within the LLNL/UCLA climate model. We discuss in detail the physical formulation, computational implementation, and application of codes for line-by-line opacity calculations and generation of wide-band opacity probability distributions. In treating the wide-band opacity probability distributions, we derive the equations for weighting the opacity probabilities of individual wavenumber bins by the Planck function. Finally, we verify our implementation of CH₄ and N₂O absorption with the Chou and Suarez model by comparison of radiative fluxes and tropospheric forcings with benchmark cases from ICRCCM and IPCC studies. Our parameterizations of the tropospheric forcing of ambient CH₄ and N₂O individually, in the absence of other trace gases, were within 13% of the ICRCCM medians. The combined forcing for the two gases was within 10% of the ICRCCM median. Our tropospheric radiative forcings for anthropogenic perturbation scenarios from IPCC and other sources compared within 2–11% with the benchmark values.

1. INTRODUCTION

Mitigation of anthropogenic global change requires developing an understanding of the scientific and economic issues involved in reducing greenhouse emissions. In the past, all greenhouse emissions have generally been treated as if they were emissions of a radiatively equivalent amount of carbon dioxide (CO₂). Radiative equivalence, in this sense, means that the same change in net radiative forcing at the tropopause is produced prior to any feedback responses. The research documented in this report is part of a project to investigate the extent to which methane (CH₄) and nitrous oxide (N₂O) have forcing and climate response patterns distinguishable from those of radiatively equivalent CO₂. The physical bases for such differences include differing spectral characteristics and spatial and seasonal distributions.

Specifically, this report summarizes the tools and methods used to parameterize the radiative absorption by CH₄ and N₂O within the context of the infrared radiative transfer model of Chou and Suarez (1994) currently being used within the LLNL/UCLA climate model. The Chou and Suarez model defines the bands and absorber contributions shown in Table 1. In adding new absorbers to the model, we have stayed within the methodology and band structure of the original model. Thus, we have added absorption by CH₄ and N₂O as also shown in Table 1. Absorption by CH₄ in band 5 and by N₂O in band 7 were evaluated but not included.

Together, they were found to contribute less than 0.005 W m^{-2} out of approximately 12 W m^{-2} to tropospheric forcing for ambient concentrations without other trace gases (see Table 3; Case 37G). In adding CH_4 absorption to band 6, it was necessary to include a semi-empirical treatment of the correlation in the overlapping with H_2O . This is also considered below.

As discussed in Chou and Suarez (1994), depending on the band and the accuracy option chosen, one of two different methods may be used to compute the transmission between two pressure levels. In cases where accurate cooling rates in the middle atmosphere (pressures less than 10 hPa) are not required, computational efficiency is increased by use of a pressure scaled k -distribution algorithm. For applications in which greater accuracy is required, the slower use of pre-computed transmittance tables combined with two-parameter (p , T) scaling is done. Our implementation of CH_4 and N_2O absorption has used the latter approach. We discuss this in more detail below.

In brief, the development of the new absorption parameterizations for the bands and gases shown in Table 1 contained the following steps. First, line-by-line opacity calculations were done at 21 pressures between 0.01 and 1000 hPa at temperatures of 170 K, 250 K, and 330 K. Depending on the pressure, gas, and band the spectral resolution for these calculations varied between $0.0005\text{--}0.01 \text{ cm}^{-1}$. Next, the line-by-line opacities were binned into Planck-weighted opacity probability densities. The probability densities were then integrated and thinned to provide cumulative probability distributions of the opacities. These distributions were then used to calculate transmissions for 21 different absorber amounts for each combination of temperature and pressure. In calculating these transmissions, a 16 stream integration over propagation angle was done, assuming isotropic radiation intensities. Finally, a quadratic expansion for the temperature was done, reducing the transmissivities for each band and gas combination to a two-dimensional table depending on pressure and absorber amount for three temperature coefficients. We discuss the tools and concepts used to implement these steps in detail below. Tests done to validate the new parameterizations against published benchmark calculations are also presented.

Table 1: Spectral bands and absorbers in the Chou and Suarez infrared model

Band	Spectral Range (cm^{-1})	Absorbers in the original model	New absorbers
1	0–340	H_2O line	
2	340–540	H_2O line	
3	540–800	H_2O line, H_2O continuum, CO_2	N_2O
4	800–980	H_2O line, H_2O continuum	
5	980–1100	H_2O line, H_2O continuum, O_3	
6	1100–1380	H_2O line, H_2O continuum	CH_4 , N_2O
7	1380–1900	H_2O line	CH_4
8	1900–3000	H_2O line	

2. CORKI — A Correlated- k Implementation

CORKI is both a specific model and an underlying conceptual framework for a set of modules that in unison create a significant infrared atmospheric radiative transfer capability. The intent of the CORKI framework has been to create a set of radiative tools to progress from line-by-line spectral properties to either detailed stand-alone radiative transfer calculations or radiative parameterizations to be included in climate and

chemistry models. CORKI is a replacement for an earlier set of models used in our previous studies such as Grossman and Grant (1994a). The development of CORKI was motivated by the need for a set of radiative transfer tools that met needs for efficiency, maintainability, and platform independence. Basic elements of the CORKI structure (Figure 1) include:

- HITRAN-96: a state of the art library of the spectroscopic properties of radiatively important atmospheric molecules. With the 1996 release, HITRAN includes HAWKS (HITRAN Advanced Work-Station). HAWKS facilitates selection and previewing of specific lines from within the entire database.
- KGEN: a moderately parallelized model for calculation of line-by-line opacities
- WC: an opacity post-processor to add the relatively smoothly varying water vapor continuum to line-by-line opacities from KGEN. The continuum opacities depends on layer temperature, pressure, and water vapor mixing ratio, as well as the wavelength. Because the Chou and Suarez infrared model already contained a treatment of continuum absorption, WC was not used in the current application.
- ABSORT: a program to calculate Planck weighted k-distributions from opacities generated by KGEN, and WC.
- TRANSMISS: a program to calculate tables of transmissivities for specific absorber amounts, pressures, and temperatures using k-distributions generated by ABSORT. Such transmissivities are then usable within the less detailed infrared radiative transfer models typical of a climate or atmospheric chemistry model.
- CORKI: a model to do accurate radiative transfer calculations for a vertically inhomogeneous atmosphere, directly using the correlated k-distribution technique. The term correlation in this application refers to the assumption that the mapping between wavenumber bins and opacity bins is approximately independent of altitude. If the opacity was factorable into a wavelength dependent absorption

Figure 1: Structural overview of the Correlated- k Implementation (CORKI) code suite for radiative transfer calculations and parameterizations.

at a specified pressure and temperature and a function of pressure and temperature alone, then the correlation would be exact. In an actual vertically inhomogeneous atmosphere, the correlation assumption is an approximation. It works well in practice, partially because the radiation field in any layer depends strongly on near-neighbor layers with which the vertical correlation is high. This radiative transfer approach was not used for the current project, and will not be further pursued in this report. We have previously discussed it in Grant et al. (1992) and Grossman and Grant (1994a).

3. KGEN

3.1 Introduction

KGEN is a moderately parallelized opacity model developed to provide a core capacity for line-by-line atmospheric radiative transfer calculations at LLNL. As opposed to most existing line-by-line codes, KGEN has been designed specifically to run on symmetric multi-processors or workstation clusters.

Calculation of line-by-line opacities from spectroscopic data is the basic foundation of all atmospheric radiative transfer problems that include gaseous absorption and emission. The applications of such opacity calculations include: development of efficient radiative parameterizations for modeling the climatic and chemical effects of potential changes in trace gases, aerosols, and cloudiness, remote sensing, and visibility calculations. Remote sensing applications include both scientific and national security aspects pertaining to the effects of atmospheric constituents on specific sensor wavelengths.

By their very nature, line-by-line opacity calculations are computationally intensive. At atmospheric pressures below about 10 hPa, integration over spectral line-shapes requires a full Voigt treatment. Simultaneously, the half-width of the line becomes significantly narrower than at larger pressures. For the 50 wavenumber interval over which the far wings of a line are assumed to have influence, there may be over 250,000 spectral bins. More over, each bin may include a quadrature of the Voigt function (Lether and Weston, 1990), resulting in over a million Voigt function evaluations per spectral line. The magnitude of this problem is multiplied by the number of lines in a desired band, often 2,000 to 12,000. Additionally, a library of spectral calculations for a particular wavenumber band and gas may require calculations at 3 separate temperatures and 26 separate pressure levels. These computational requirements have here-to-fore limited such codes to vector supercomputers.

We have used the KGEN opacity model to generate correlated-k (opacity probability) distributions for computationally efficient solar and terrestrial infrared radiative transfer calculations. Such k-distributions can be used directly for detailed radiative transfer calculations or be used to tabulate wide-band spectral atmospheric transmission as a function of path absorber amount, effective path pressure, and effective path temperature. Such wide-band transmission data are the basis for efficient interactive radiative transfer calculations within climate models. We calculated a library set of opacities for CH₄ and N₂O executing KGEN on a cluster of DEC Alpha workstations.

3.2 Description of Computational Methods

The absorption coefficient of a molecule at a particular wavenumber ν (cm⁻¹) and temperature is given by the product of the temperature dependent line strength $S(T)$ and the line shape factor $f(\nu - \nu_0)$, where ν_0 is the position of the line center. Including the normalization of the line, we have

$$\begin{aligned} k_\nu &= S(T)f(\nu - \nu_0) , \\ S &= \int_{-\infty}^{\infty} k_\nu d\nu . \end{aligned} \tag{1}$$

The variation of the line strength, S , with temperature is given by

$$S(T) = S(T_r) \frac{Q(T_r)}{Q(T)} \frac{1 - e^{-hc\nu_0/KT}}{1 - e^{-hc\nu_0/KT_r}} \exp\left[-\frac{hcE_i}{K} \left(\frac{1}{T} - \frac{1}{T_r}\right)\right], \quad (2)$$

where T_r is a reference temperature, E_i is the energy of the lower state of transition, and $Q(T)$ is the internal partition function for the molecule in question (Liou, 1992). The evaluation of Eqn. 2 is facilitated by $S(T_r)$ and E_i being part of the spectral data provided for each line in the HITRAN database. A function for the evaluation of $Q(T_r)/Q(T)$ based on quadratic fits has also been included in the HITRAN release (Gamache et al., 1990).

The line shape factor is given by

$$f_\nu(\nu - \nu_0) = \frac{1}{\alpha_D \sqrt{\pi}} K(x, y), \quad (3)$$

where α_D is the Doppler width (Liou, 1992). Further defining α to be the Lorentz half-width; $y = \alpha/\alpha_D$, and $x = (\nu - \nu_0)/\alpha_D$, the Voigt function is defined by

$$K(x, y) = \frac{y}{\pi} \int_{-\infty}^{\infty} \frac{1}{y^2 + (x - t)^2} e^{-t^2} dt. \quad (4)$$

In the lower atmosphere where the Lorentz half-width is much greater than the Doppler width, the line shape simplifies to the Lorentz profile. In the upper atmosphere, where the Lorentz width is negligible, the line shape simplifies to the Doppler profile. In between these extremes, the more complex Voigt profile must be used.

We perform the many Voigt function evaluations required using the method of Lether and Wenston (1990). This method is based on a corrected trapezoidal integration in which the number of terms can be varied to trade-off computational speed and accuracy. Lynas-Gray (1993) had subsequently implemented this algorithm for vector processors. Continued KGEN development efforts will include additional optimization of our application of this algorithm as well as further development of post-processing, testing, and analysis tools. Output from the KGEN model is to NetCDF files, facilitating platform portability of the data, self-documentation, and ease of further analysis.

The KGEN model has been parallelized and tested on a DECAAlpha cluster using the Message Passing Interface (MPI) described by Gropp et al. (1994). The accuracy of the code was verified against a previous model (Grossman and Grant, 1994). The trapezoidal scheduling algorithm of Tzen and Ni (1993) was implemented to load balance the calculation of opacities in the different spectral bins affected by each spectral line. We have verified linear increases in through-put for one to five slave processors on ten CPU (central processing unit) machines. For a larger number of processors, competition with other tasks resulting in thrashing delays, resulting in a noticeable decrease in computational efficiency. Avoidance of such tasking conflicts requires gang scheduling of CPUs.

4. ABSORT

4.1 Background

In a paper by Lacis and Oinas (1991), a method was described for treating transmission calculations in infrared radiative transfer problems for a vertically inhomogeneous atmosphere in which there is non-gray gaseous absorption. The technique, known as the correlated k -distribution method, utilizes a mapping of the

opacity-frequency relation into an opacity-probability relation within a particular frequency interval. We have previously summarized the basic theory and some results of applying the method in Grossman and Grant (1994a). The cumulative probability distribution, used in defining the “c-k” method is given by

$$G(k) = \int_{-\infty}^k f(k') d \log k' , \quad (5)$$

where $f(k') d \log k'$ is the fraction of the fraction of the frequency interval occupied by absorption coefficients between $\log k'$ and $\log k' + d \log k'$, i.e. the probability density function. Using these definitions, the transmission through a single, homogeneous layer can be expressed in three equivalent forms:

$$\begin{aligned} T(u) &= (\Delta v)^{-1} \int_{\Delta v} e^{-k(v)u} dv , \\ &= \int_{-\infty}^{\infty} f(k) e^{-ku} d \log k , \\ &= \int_0^1 e^{-k(G)u} dG , \end{aligned} \quad (6)$$

where u is the path absorber amount. The advantage of making these transformations is in going from an integral in which k is rapidly varying over orders of magnitude to one in which k is monotonically and smoothly increasing. The properties of smoothness and monotonicity yield themselves to efficient numerical evaluation of the integral. The term “correlated” refers to the assumption that, for a vertically inhomogeneous atmosphere, the mapping or ordering relating wavelength bins to opacity bins will remain constant. For any real atmosphere this is an approximation. It holds exactly under the assumption the wavelength variation and pressure-temperature variation of the opacities can be separated into independent factors.

Now consider an infrared band of sufficient width that not only the variation in the opacity but also the variation in the Planck function must be considered. We can sub-divide such a band, as has been done by Chou and Suarez (1994), so that within each sub-band the Planck function could be taken as constant. We can then write the Planck-weighted total band transmission as the sum over the sub-bands given by

$$T(u) = \sum_{j=1}^J \frac{B_j(T)}{B(T)} \Delta v_j \int_0^{\infty} e^{-ku} f_j(k) d \log(k) , \quad (7)$$

where $B(T) = \sum_j B_j(T) \Delta v_j$ is the band-integrated Planck flux, $B_j(T)$ is the Planck flux in the j th sub-band, and $f_j(k)$ is the probability density function of the absorption coefficient in the j th sub-band. Discretizing the integral over k in Eqn. 7 into a sum over N terms and rearranging, we obtain

$$\begin{aligned} T(u) &= \sum_{n=1}^N e^{-k_n u} \left[\Delta \log(k_n) \sum_{j=1}^J \frac{B_j(T)}{B(T)} \Delta v_j f_j(k_n) \right] , \\ &= \sum_{n=1}^N e^{-k_n u} \Delta \tilde{G}_n , \end{aligned} \quad (8)$$

with $\Delta \tilde{G}_n$ interpreted as being a Planck-weighted probability increment or density. If we now go to the limiting case of the sub-bands being single wavelength bins, the unweighted probability density terms $f_j(k_n)$ become delta functions, since the opacity for a particular wavelength bin will lie in one and only one opacity bin. In this case, we can express $\Delta \tilde{G}_n$ by

$$\Delta \tilde{G}_n = \left[\Delta \log(k_n) \sum_{j=1}^J \frac{B_j(T)}{B(T)} \Delta v_j \delta(k_j - k_n) \right] G_{\infty}^{-1} , \quad (9)$$

saving the necessity of an intermediate step of calculating separate k -distributions for each sub-band in transforming from line-by-line opacities to a total band k -distribution. The normalization factor, G_∞^{-1} , is defined such that $\sum_n \Delta \tilde{G}_n = 1$. Examples of the resulting opacity Planck-weighted probability density function and cumulative probability distribution are shown in Figures 2 and 3.

The practical effect of adding the Planck weighting is to give unequal weighting to the different wavelength bins in calculating the probability density function. Specifically, as discussed by Chou and Suarez (1994) and Liou (1992), the terms defining the interaction of the radiation field with a discrete atmospheric layer are differences of individual terms of the form

$$Q = \sum_{j=1}^J B_j(T) \Delta v_j e^{-k_j u}, \quad (10)$$

where the sum is taken over the individual wavenumber bins. In this form, it is readily apparent that the weight given to each exponential transmission involving k_j is given by $B_j(T) \Delta v_j$. By mapping the weights of individual wavenumber bins into the N discrete opacity bins $\Delta \log(k_n)$, $n = 1, 2, 3, \dots, N$, and subsequently renormalizing, we arrive again at the result of Eqn. 9.

Rather than assuming $B_\nu(T)$ to be constant over some sub-band or evaluating it in full for each of many wavelength bins, we can evaluate it at some subset of wavelengths and interpolate in-between. In practice, the variation in the shape of the Planck function over the range of atmospheric temperatures is of far less importance than its original inclusion. Chou and Suarez (1994) have noted that, for the weighting purposes described above, the evaluation of the Planck weighting at a temperature of 250 K is accurate for all atmospheric use.

4.2 Implementation Issues

At low pressures, the centers of strong absorption lines are not appreciably broadened from their doppler limits. The effect of this on the opacity probability distributions is to create a small but optically significant probability for opacities relatively large compared to the remainder of the distribution. This results in a sharp peak in the opacities for cumulative probabilities between 0.99 and 1. In choosing the values of cumulative probability, G , for the final k -distribution $k(G)$, accuracy requires care in resolving this peak.

Figure 2: Planck-weighted probability density functions for CH_4 opacities in band 6 at a pressure of 0.01 hPa and at temperatures of 170 K, 250 K, and 330 K.

Figure 3: Planck-weighted cumulative probability distributions for CH_4 opacities in band 6 at a pressure of 0.01 hPa and at temperatures of 170 K, 250 K, and 330 K.

Table 2: The first values, intervals, and sizes of the pre-computed tables of the regression coefficients a , b , and c of Eqn. 13 for transmittance calculations. The units of u for both CH_4 and N_2O are g cm^{-2} . The units of pressure are hPa.

Absorber	$\log_{10}(u)_1$	$\Delta\log_{10}(u)$	u table size	$\log_{10}(p_e)_1$	$\Delta\log_{10}(p_e)$	p_e table size
CH_4	-8.0	0.3	21	-2	0.2	26
N_2O	-8.0	0.3	21	-2	0.2	26

In creating the probability distribution $f(k)$ from $k(v)$, we may start with mapping several hundred thousand wavelength bins into several thousand k -distribution bins. The k -distribution bins are most often chosen to have equal increments in $\log(k)$. In order to use the c-k method for atmospheric modeling, we must establish the necessary resolution in G at the top of the atmosphere where the resolution requirements are most stringent. The same set of G 's must then be used for all vertical layers below. To facilitate this, we fit the relation

$$G(\hat{k}) = 1 + e^{-a\hat{k}^b} - \hat{k}e^{-a}, \quad (11)$$

for the coefficients a and b with equally spaced $\hat{k}$ defined by

$$\hat{k} = \frac{\log(k) - \log(k_{\min})}{\log(k_{\max}) - \log(k_{\min})}. \quad (12)$$

Given the number of G points desired and the coefficients a and b , the cumulative probability discretization is uniquely determined. This provides a compact method of specifying a particular G discretization to ABSORT for analyses in which the output gridding has already been determined from a prior analysis. Having calculated the cumulative probability distribution for a layer at specified points in $\log(k)$, the inversion to $\log(k(G))$ at specified G points is accomplished via piecewise cubic Hermite interpolation.

5. TRANSMISS

TRANSMISS accomplishes the final step in going from spectroscopic data tables to tables of transmission for specified absorber path amounts, pressures, and temperatures. To implement absorption by CH_4 and N_2O within the model of Chou and Suarez (1994), path transmittances for each gas within each affected wavelength band were calculated at 26 pressures, 21 absorber amounts, and 3 temperatures. Calculations were done at temperatures of 170 K, 250 K, and 330 K. Because the temperature dependence of the transmittances is smooth and relatively slowly varying, the table look-up can be reduced from three-dimensional to two-dimension using a quadratic fit in temperature. We then have,

$$T(u, p_e, T_e) = a(u, p_e) + b(u, p_e)(T_e - 250 \text{ K}) + c(u, p_e)(T_e - 250 \text{ K})^2 \quad (13)$$

where u is the path absorber amount and p_e and T_e are absorber-weighted effective pressures and temperatures for a given path. Values of flux transmittances are tabulated for the pressures and absorber amounts given in Table 2. Because the tables of transmittances are precomputed, it was not necessary to use a single effective propagation angle, or equivalently a diffusivity factor to achieve computational efficiency. We therefore used a 16 stream angular integration assuming isotropic intensity in calculating the final flux transmissivities.

6. Using the Parameterization

The transmissivity data for CH₄ and N₂O is implemented within the Chou and Suarez model as transmissivities at a temperature of 250 K and linear and quadratic temperature expansion coefficients. For each band and gas, the data are provided at the pressures and absorber amounts given in Table 2. Temperatures within the model are handled internally as deviations from 250 K.

For each absorber in the model, the column integrated amount from the top of the atmosphere to each level (layer boundary) is calculated. The absorber-weighted integrals of pressure and temperature deviation are also calculated. When a transmissivity between two levels is needed, the absorber path-amount, and effective average pressure and temperature deviation are thus readily available. Given these three parameters, the model locates the bounding pressures and absorber amounts in the transmissivity table, and interpolates the transmissivity coefficients to the current pressure and absorber. The quadratic expansion is then made to evaluate the needed transmissivity.

The overlapping between successive absorbers is handled via the transmission product rule. If we denote the mean flux transmission of the j 'th gas over a band as T_j and the deviation from the mean as T_j' , then we can write the overlapped transmission T_{12} of two gases as

$$T_{12} = T_1 T_2 + \frac{\int_{\Delta\nu} B_\nu T_1'(\nu) T_2'(\nu) d\nu}{\int_{\Delta\nu} B_\nu d\nu}, \quad (14)$$

where the first term on the right constitutes the standard product rule and the second term is nonzero only if the variation of transmissivity of the two gases is spectrally correlated. In testing the parameterization of CH₄ in band 6, we discovered that, while the absorption by CH₄ alone or overlapped only with N₂O seemed to be correct, the absorption overlapped with H₂O was too large. For example, in a test of the forcing due to the 1990–1995 increase in CH₄ with other gases held constant, we initially calculated a forcing of 0.71 W m⁻² rather than the 0.52 W m⁻² of our benchmark calculation. This discrepancy is explainable as the result of ignoring a positive correlation in Eqn. 14. In treating the overlap of CO₂ and H₂O in band 3, Chou and Suarez (1994) had solved the problem by dividing the band into three sub-bands. However, in making our modifications to the original model, we did not want to change the existing parameterization of H₂O in band 6. We solved this by using a semi-empirical parameterization for the overlap correlation having the correct limiting properties and having one parameter tuned to our ambient benchmark forcing studies. The overlap parameterization we used is given by

$$T_{12} = T_1 T_2 \left[1 + \gamma (1 - T_1) (1 - T_2) \frac{p}{p_0} \right], \quad (15)$$

with γ equal to 1.85.

To validate our implementation of CH₄ and N₂O absorption, we have compared our results for a number of radiative flux and forcing calculations with those from other studies. In Table 3, we compare our results of flux calculations with those from the Intercomparison of Radiation Codes use in Climate Models (ICRCCM) presented by Ellingson et al. (1991). Comparisons are at the surface tropopause, and the top of the atmosphere. These results, being for CH₄ and N₂O only, are unaffected by the overlap correlation correction of Eqn. 15. They are thus a direct measure of the accuracy of the new parameterizations independent of other absorbing gases.

Comparing our results with ICRCCM results in Table 3, our parameterizations of the tropospheric forcing of ambient CH₄ and N₂O individually, in the absence of other trace gases, were within 13% of the ICRCCM medians. These forcing estimates are based on an upward surface flux of 423 W m⁻² for the mid-latitude

summer surface temperature of 294 K. We would expect the medians results to be less influenced by outliers than the means. The combined forcing for the two gases was within 10% of the ICRCCM median.

In Table 4, we compare radiative forcing calculated at the tropopause for a clear-sky mid-latitude summer atmosphere and a global average atmosphere, including clouds. Radiative forcing cases were selected from several sources, including the International Panel on Climate Change Second Assessment Report (IPCC, 1995). In all test cases, the results of our parameterizations for CH₄ and N₂O absorption are within acceptable ranges of variability (2–11%) as compared with other studies. While the band 6 transmission overlap of CH₄ with H₂O was corrected using Eqn. 15, the same correlation strength parameter was used for all calculations. The consistent agreement over all cases, without further tuning, is an indication that the use of Eqn. 15 is robust.

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Table 3: Comparison with ICRCCM infrared fluxes for CH₄ and N₂O cases

ICRCCM Case	New Parameterization (W m ⁻²)	ICRCCM			
		Median (W m ⁻²)	Range (W m ⁻²)	Mean (W m ⁻²)	Std. Dev. (W m ⁻²)
37A (11) — 1.75 ppmv CH ₄					
Surface	417.19	417.06	7.73	415.77	2.20
Tropopause	419.37	418.76	3.86	418.23	1.13
TOA	419.25	418.55	3.84	417.96	1.14
37B (9) — 0.28 ppmv N ₂ O					
Surface	417.13	417.22	8.24	415.70	2.69
Tropopause	419.68	419.13	5.15	418.57	1.44
TOA	419.33	418.58	5.38	418.06	1.58
37G (10) — CH ₄ & N ₂ O					
Surface	411.26	411.64	14.71	409.18	4.58
Tropopause	415.71	414.91	11.05	413.63	3.11
TOA	415.28	414.29	15.05	412.66	4.14

Table 4: Comparison with MLS, global average, and IPCC forcing cases

Case	New Parameterization (W m ⁻²)	Comparison 1 (W m ⁻²)	Comparison 2 (W m ⁻²)
MLS — Clear Sky No other gases present CH ₄ : 1.72 – 3.44 ppmv	1.63	Grossman & Grant (1994b) 1.67	
MLS — Clear Sky Ambient trace gases CH ₄ : 0 – 1.72 ppmv	1.81	Grossman & Grant (1994b) 1.69	Ramaswamy (1993) 2.02
MLS — Clear Sky Ambient trace gases CH ₄ : 1.72 – 3.44 ppmv	0.769	Grossman & Grant (1994b) 0.718	Shine (1993) 0.852
Global Average Atmos. 1990 Ambient Gases CH ₄ : 1.65 – 3.30 ppmv	0.527	Grossman & Grant (1994b) 0.521	Ramanathan et al. (1987) 0.550
IPCC 1990 – 1995 1990 Ambient Gases N ₂ O: 0.310 – 0.314	0.015	IPCC 0.016	
IPCC 1990 – 2000 1990 Ambient Gases N ₂ O: 0.310 – 0.319	0.032	IPCC 0.033	

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