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A.S. Grossman
K.E. Grant
D.J. Wuebbles

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Radiative Forcing Calculations for CH_3Cl

Allen S. Grossman
Keith E. Grant
Donald J. Wuebbles*

Global Climate Research Division, L-262
Lawrence Livermore National Laboratory
P.O. 808, Livermore CA 94551
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Abstract

Methyl chloride, CH_3Cl, is the major natural source of chlorine to the stratosphere. The production of CH_3Cl is dominated by biological sources from the oceans but it also has smaller anthropogenic sources, such as biomass burning. Production has a seasonal cycle which couples with the short lifetime of tropospheric CH_3Cl to produce nonuniform global mixing. As an absorber of infrared radiation, CH_3Cl is of interest for its potential affect on the tropospheric energy balance as well as for its chemical interactions. In this study, we estimate the radiative forcing and global warming potential (GWP) of CH_3Cl. Our calculations use an infrared radiative transfer model based on the correlated k-distribution algorithm for band absorption. A radiative forcing value of 0.0053 W/m2 /ppbv was obtained for CH_3Cl and is approximately linear in the background abundance. This value is about 3 percent of the forcing of CFC-11 and about 300 times the forcing of CO_2, on a per molecule basis.The radiative forcing calculation for CH_3Cl is used to estimate the global warming potential (GWP) of CH_3Cl . The results give GWPs for CH_3Cl of about 30 at a time of 20 years($CO_2 = 1$). This result indicates that while CH_3Cl has a GWP similar to that of CH_4, the emission rates are too low to meaningfully contribute to atmospheric greenhouse heating effects.

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* now at Dept. of Atmospheric Sciences, Univ. of Illinois, Urbana, Ill., 61801

I. Introduction

Methyl chloride, CH_3Cl, is the most abundant halocarbon in the earth's atmosphere (Elkins et al., 1984), representing approximately 30 percent of the total chlorine content. Typical concentrations on the order of 600 parts per trillion have been determined (Rasmussen et al., 1980, Singh et al., 1983, WMO, 1991, Kaye et al., 1994). Volconic activity, biological production, automobile exhaust, and biomass burning have been considered to be CH_3Cl sources (Crutzen et al. 1979). The total budget is currently dominated by natural sources. The principal sink for CH_3Cl is the reaction with OH in the troposphere (Howard and Evenson, 1976). The atmospheric lifetime for CH_3Cl is 1.5 years, (Prather, 1989, WMO, 1991, 1994). According to Elkins et al. (1984), CH_3Cl has strong infrared absorption bands at 732, 1015, 1355, 1455, 2879, 2966 and 3042 cm^{-1}. As a result of these strong absorption bands the potential exists for possible, anthropogenically based, greenhouse heating of the atmosphere that could result in climate changes.

The mechanism usually used for comparison of the greenhouse potential of trace gases is the global warming potential or GWP as defined by IPCC (1990). The GWP is the ratio of the time integrated radiative flux change at the tropopause caused by the introduction of a unit mass impulse of a trace gas into the atmosphere to the time integrated radiative flux change at the tropopause caused by the introduction of a unit mass impulse of CO_2. An essential part of the GWP determination is the calculation of the radiative forcing, which is defined as the radiative flux change at the tropopause produced by a unit change in the number of molecules of a particular gas with all other abundances held constant. Usually the amount of abundance change used in a radiative forcing calculation is chosen such that the change is just enough to produce a numerically significant flux change value at the tropopause. Parameterized experiments for radiative forcing have been published, for example, in IPCC (1990, 1992, 1994), and by Ramanathan et. al. (1987). A detailed radiative transfer model is required in order to calculate the radiative forcing for the GWP determination. Grant et al. (1992, "GGFP") and Grossman and Grant (1992a) have used a correlated k-distribution model for the absorption by the major atmospheric molecular absorption species (H_2O, CO_2, O_3, CH_4, and N_2O) to calculate the fluxes and heating rates in the 0–2500 cm^{-1} wavenumber range. The fluxes and heating rates obtained for this model are accurate to well within ten percent when compared to line by line calculations. The altitude range covered by these calculations was 0–60 km.

The main purpose of this paper is to calculate the tropospheric radiative forcing of CH_3Cl using the correlated k-distribution radiative transfer model and the line by line data in the HITRAN91 data base (Rothman et al., 1991) and the spectroscopic line data of Brown (1994). The calculation will be done for a globally and annually averaged model atmosphere with a representative cloud distribution. GWP calculations for CH_3Cl will be made and compared to other trace gas GWPs given in IPCC (1992, 1994).

II. Global Warming Potential

The GWP of a gas is defined as,

$$GWP(c_i) = \frac{\int_0^t \Delta F(c_i, t)dt}{\int_0^t \Delta F(c_{CO_2}, t)dt} \quad , \tag{1}$$

where ΔF is the change in radiative forcing with time as a function of the species concentration (c_i). An approximate form of Equation 1 was published in IPCC (1990) and is given by the expression;

$$\mathrm{GWP}\,(c_i) = \frac{\int_0^t a_i c_i dt}{\int_0^t a_{co_2} c_{co_2} dt} \quad , \tag{2}$$

where a_i is the instantaneous radiative forcing (per unit mass) due to a unit increase in the concentration of trace gas i and ci is the concentration of the trace gas i remaining at time t after its release. The corresponding values for carbon dioxide are in the denominator. In the discussion below we describe a technique for approximating the direct GWP for any greenhouse gas relative to the GWP for CCl_3F (CFC-11). This technique follows directly from the IPCC definition for GWPs. This approximation assumes that the emission impulse is small enough that the radiative forcing of c_i is linear with concentration. The GWP of a gas c_i referred to as (GWP(c_i)) can be expressed as,

$$\mathrm{GWP}(c_i) = \frac{\int_o^t a_i c_i dt}{\int_o^t a_{CFC-11} c_{CFC-11} dt} \cdot \mathrm{GWP(CFC\text{-}11)}. \tag{3}$$

The quantity c_i can be approximated by the relation,

$$c_i = co_i \ \exp\left(-t / \tau_i\right), \tag{4}$$

and Equation 3 becomes,

$$\mathrm{GWP}\ (c_i) = \frac{\int_o^t a_i co_i \exp\left(-t / \tau_i\right) dt}{\int_o^t a_{CFC-11} co_{FC-11} \exp\left(-t / \tau_{CFC-11}\right) dt} \cdot \mathrm{GWP(CFC\text{-}11)}. \tag{5}$$

Assume a_{Ci4} and $a_{Ci\text{-}11}$ are constant and, by definition, $co_{Ci4} = co_{CFC\text{-}11}$ (since both assume the same mass emission impulse into the atmosphere). Equation 5 becomes

$$GWP(C_i) = \frac{m_{CFC-11}}{m_i} \cdot \frac{a_i}{a_{CFC-11}} \cdot \frac{\tau_i}{\tau_{CFC-11}} \cdot \frac{\left(1 - \exp(-t/\tau_i)\right)}{\left(1 - \exp(-t/\tau_{CFC\text{-}11})\right)} GWP(CFC - 11) \tag{6}$$

where the a's are the radiative forcing values at the tropopause in W/m^2 per ppbv, the m's are the molecular mass, and the τ's are the atmospheric lifetimes. The radiative forcing term a_i in Equation 2 is defined as the difference in the net radiative flux at the tropopause due to a change in the composition by 1 ppbv of a single molecular species while, at the same time, keeping the composition of all other species constant.

As a test of the approximate model, the GWP of CCl_2F_2 (CFC-12) is calculated and compared to the values given in IPCC (1992). Equation 6 is used to calculate the GWP(CFC-12) for times of 20, 100, and 500 years and the results are given along with the values published in IPCC, (1992) and the percentage of error in Table 1.

Table 1. GWP Model Comparison

GWP (CO_2=1)				
		Time (years)		
Gas	Lifetime	20	100	500
CFC-11	50	4500	3400	1400
CFC-12 (IPCC, 1992)	116	7100	7100	4100
CFC-12 (derived)		7152	7174	4226
% error		0.7	1.0	3.1

III. Correlated K-Distribution Radiative Transfer Model

The correlated k-distribution method utilizes a mapping of the absorption coefficient vs. wavenumber relation into an absorption coefficient vs. probability relation within a particular wavenumber interval. The probability variable g(k), the cumulative distribution function, is defined as

$$g(k) = \int_0^k f(k')\,dk' \ , \tag{7}$$

where f(k')dk' is the fraction of the frequency interval occupied by absorption coefficients between k' and k'+dk'(Goody and Jung, 1989, "G1"; Goody et al., 1989, "G2"; and West et al., 1990, "W1"). The limits of g(k) range between 0 and 1 within the frequency interval. The inverse of Equation 7, k(g), the k-distribution, has been shown by G2, and W1 to be a monotonic function across the frequency interval for a particular atmospheric layer. The correlated k-distribution method can mathematically provide an exact procedure for calculating the transmission, fluxes, and heating rates in a homogeneous atmosphere. For the case of inhomogeneous atmospheric paths the method is, in practice, inherently inexact since somewhat different sets of frequencies will associate with a given ordering of the k terms as the pressure and temperature vary over the path. Numerous tests of the model for various atmospheric trace gases and atmospheric temperature - pressure profiles, Grossman and Grant (1992a, b, 1994) and Grossman et al. (1993), show that the method produces fluxes that are accurate to well within ten percent when compared to line by line calculations.

The calculation of the transmission can be expressed in the three physically equivalent forms:

$$\begin{aligned} T(u) &= 1 / \Delta\upsilon \int_{\Delta\upsilon} \exp\left(-k_{\upsilon} u\right) d\upsilon \quad , \\ &= \int_{0}^{\infty} f(k') \exp(-k' u) dk' \quad , \\ &= \int_{0}^{1} \exp\left(-k(g) u\right) dg \quad , \end{aligned} \tag{8}$$

where u is the absorber column density. Using the k -distribution form, the calculation can be performed with far fewer k-g points than the same calculation using k-wn (wavenumber) points.

The direct calculation of the molecular k-distributions contains the following steps (GGFP). First the HITRAN database (Rothman et. al., 1991) is utilized to determine the line transitions and physical properties of the selected lines. These line properties are merged with the line properties of CH_3Cl provided in the data base of Brown (1994) to give a complete set of CH_3Cl lines. Second, a modified version of the FASCODE2 code (Clough et. al., 1986) is used to calculate a finely gridded set of monochromatic absorption coefficients, with full allowance for the overlap of neighboring lines, for each layer in the atmosphere. Third, a sorting code, ABSORT, is used to calculate the f(k), g(k), and k(g) functions for each homogeneous layer. The modified FASCODE program takes the line data and fits an absorption line profile (Voigt profile) to each line and calculates the absorption coefficient k (cm^2/air mol) as a function of wavenumber. The normal cutoff point in the line profile is set at 25 cm^{-1} from line center. This is done for reasons of economy (beyond 25 cm^{-1} a given line contributes little absorption). The ABSORT code takes the absorption coefficient files generated by the FASCODE program and sorts the absorption coefficients into bins of equal logarithmic width, $\Delta \log k$, to produce a distribution function, f(k), based on the relative probability of occurence within the wave number interval (proportional to the number of entries in each bin). The cumulative distribution function, g(k) (c.f. Equation 7), is obtained by numerical integration of the f(k) function. The k-distribution, k(g), is obtained by a reverse interpolation of the g(k) relation using a spline function. For the calculations in this paper a 401 bin model was used to insure an adequate number of points at g values between 0.9 and 1.0. This g value region is important for heating rate calculations at high altitudes. The output from ABSORT is the 401 point k(g) relation for each layer. At low pressures the k(g) curves can show opacity variations of up to five orders of magnitude at g values greater than ~0.9. This kind of behavior at low pressures is thought to be due to the absence of pressure broadening on the absorption lines in the wave number band; i.e. the lines are dominated by doppler broadening near line center. These variations in the k-distributions require a careful numerical integration strategy in the transmission expression, Equation 8, in order to accurately reproduce the k(g) functions. The integration strategy which was adopted after test calculations was an 85 point variable spaced trapezoidal model with g spacings of 0.0025 for g values between 0.9 and 1.0 and larger g spacings at lower g values.

IV. CH_3Cl Data

Inspection of the HITRAN91 database reveals that only the lines of the V1, V4, and 3V6 bands of CH_3Cl between 2907 and 3173 cm^{-1} have been included in the compilation. According to Elkins et al. (1984) the strength of these bands represent approximately 40 percent of the total line strength of all CH_3Cl bands. Furthermore this spectral region contains water vapor bands and the CH_3Cl contributions to the radiative forcing may be small due to overlapping water vapor absorption. A database for the properties of the CH_3CL lines in the V2, V3, and V5 bands has been developed by Brown (1994) and Brown et al. (1987) in the wavenumber regions 661 to 772 cm^{-1} (V3) and 1261 to 1646 cm^{-1} (V2, V5). This is an IR window region and thus these bands should contribute the majority of the radiative forcing. According to Elkins et al. (1984) the V2, V3, and V5 bands represent approximately 55 percent of the total line strength of the CH_3Cl bands. The V6 band at 1015 cm-1 is not presently tabulated on any database and cannot be included in the radiative forcing calculation. The effect of the V6 band omission would be on the order of 5 percent. The combined strength of the V2, V3, and V5 bands should be approximately 1.34 times the combined strength of the V1, V4, and 3V6 bands, Elkins et al. (1984). The ratio of the combined line strength of the V2, V3, and V5 bands in the Brown (1994) to the combined line strength of the V1, V4, and 3V6 bands in the HITRAN91 data base is 3.92 indicating that the HITRAN line strengths may be too low by a factor of 2.92. Radiative forcing calculations will be done for the HITRAN91 line strengths as given and for the case where the line strengths have been multiplied by a factor of 3. Both the Brown (1994) and the HITRAN91 batabases were numerically merged in the calculation of the k-distributions outlined in GGFP (1992) in order to calculate the complete radiative forcing of CH_3Cl.

V. Parameters of The Calculations

Flux and radiative forcing calculations were made for a globally and seasonally averaged model atmosphere (Wuebbles et al., 1994). The mixing ratio vs. altitude profiles for H_2O, O_3 and CH_4 are shown in Figure 1. CO_2 was assumed to have a mixing ratio of 350 ppmv, constant with altitude. N_2O was assumed to have a mixing ratio of ~0.3 ppmv in the troposphere and then decrease to a mixing atio of ~1.2 ppbv at 60 km altitude. The temperature-pressure profile for model atmosphere is shown in Figure 2. The tropopause in the globally-averaged atmosphere is specified as the altitude at which the temperature gradient in the troposphere decreases to 2K/km. This occurs at a pressure of 166 mb (~13.2 km). Altitude resolution in the model atmosphere was 1 km at altitudes between 0 and 20 km, and 2 km at altitudes between 20 and 60 km. The ground temperature was 291 K. The clear sky radiative transfer model outlined in GGFP was modified to accept a cloud distribution model using an algorithm based on Harshvardhan et al. (1987). In this algorithm the transmission between atmospheric layers is multiplied by the probability of a clear line of sight between the layers. The clouds are considered to be radiatively black at the thermal wavelengths. For the case of random overlap of the cloud layers, which is the case adopted in this paper, the probability of a clear line of sight between two layers i and j is given as,

$$C_{ij} = (1-N_{j-1})(1-N_{j-2})..........(1-N_i) \quad , \tag{9}$$

where the N's represent the fractional cloud cover of the particular layers. The transmission is then given as,

$$T_{ij} = T_{clr}C_{ij} \quad , \tag{10}$$

where T_{clr} is the clear sky transmission. The cloud distribution in the globally averaged atmosphere consists of three layers, each 1km thick, with bases at 2 km (low), 4 km (middle), and 10 km (high). Fractional cloud cover amounts are 0.31 (low), 0.09 (middle), and).17 (high). The radiative transfer calculations to determine the tropospheric radiative forcing were carried out over the wavenumber range of 500 - 3000 cm^{-1}, in 25 cm^{-1} subintervals. In addition to CH_3Cl absorption, absorption due to H_2O, CO_2, O_3, CH_4, and N_2O was included in the calculations. CH_3Cl mixing ratios of 0.0 (ambient), 1 ppbv (forced), and 100 ppbv (forced), constant with altitude were used.

VI. Results and Discussion

The tropospheric radiative forcing calculations for CH_3Cl are shown in Table 2.

Table 2. Tropospheric radiative forcing calculations for CH_3Cl.

INCLUDED BANDS	RADIATIVEFORCING(W/m^2/ppbv)
v1, v2, v3, v4, v5, 3v6 (100 ppbv forcing)	5.30e-03
v1, v2, v3, v4, v5, 3v6 (1 ppbv forcing)	6.08e-03
v1, v4, 3v6 (HITRAN91)	2.71e-06
v1, v4, 3v6) X 3	15e-06

The results of Table 2 show that the tropospheric radiative forcing of CH_3Cl is due entirely to the V2, V3, and V5 bands. The contribution of the V1, V4, and 3V6, bands, even with a factor of 3 increase in the line strengths, is most likely heavily overlapped by water vapor lines. Calculations were performed using both 1 ppbv and 100 ppbv forcing to insure that the 1 ppbv forcing result was numerically significant and to determine the linearity effects in the forcing as a result of increased CH_3Cl abundance. It appears that the radiative forcing is linear to within approximately 13 percent in the abundance of CH_3Cl between 1 and 100 ppbv. Using the radiative forcing formulae given in IPCC (1990), the radiative forcing of CH_3Cl is about 3 percent of that of CFC-11 and about 300 times that of CO_2, on a per molecule basis. CH_3Cl also has a larger radiative forcing than CH_4 (21 times CO_2) and N_2O (206 times CO_2), IPCC (1990).

Although a trace gas can have a strong radiative forcing per molecule, its greenhouse heating potential of the atmosphere depends also on the lifetime of an impulse of the trace gas to the atmosphere as well as its time dependent anthropogenic emission into the atmosphere. The GWP for the trace gas addresses the net effect of the combination of the radiative forcing and the lifetime of the gas by calculating the time integrated radiative forcing of a unit mass impulse to the atmosphere. Table 3 shows the results of a calculation of the GWP for CH_3Cl at times of 20, 100, and 500 years based on CFC-11 GWPs as determined in IPCC (1994) (c.f. Equation 6). The lifetime of CH3Cl used in the calculation was 1.5 years.

Table 3. Global warming potential for CH_3Cl.

20 YEARS	100 YEARS	500 YEARS
29.9	9.1	2.8

For CFC-11, with a lifetime of 50 years, the GWP at 20, 100, and 500 year integration periods are 5000, 4000, and 1400 respectively IPCC (1994). The GWPs of CH_3Cl given in Table 3 are approximately 37 to 48 percent of GWPs of CH_4, on a per kilogram basis (IPCC, 1994). Wuebbles et al. (1994) calculate GWP values calculate GWP values for CH_4 which are 31 to 45 percent larger than the IPCC (1994) values due to larger CH_4 response times. With regard to these larger GWP values, the CH_3Cl GWP values range from 25 - 37 percent of the CH_4 GWPs. Kaye et al. (1994) give the abundance of CH_3Cl as approximately 600 parts per trillion in the troposphere, decreasing to approximately 20 parts per trillion at altitudes around 30 km, with an annual emission of approximately 3.5 million tons per year. Kay et al. (1994) estimate an anthropogenic CH_3Cl emission rate of between 15 and 30 percent of the total emission rate, principally due to biomass burning. Given a methane emission rate of approximately 500 Tg/year (WMO, 1991), with approximately 50 percent of the total due to anthropogenic sources (IPCC,1990), the global warming effects of CH_3Cl are about 0.07 to 0.2 percent of the methane contribution contribution using IPCC (1994) methane GWP values. The global warming effects of CH_3Cl are about 0.04 to 0.14 percent of the methane contribution contribution using the Wuebbles et al. (1994) methane GWP values. Thus, at present, serious greenhouse problems are not a current problem and will not become a problem unless very large anthropogenic releases of this gas occur.

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Figure Captions

Figure 1. Globally and annually averaged profiles of water vapor, ozone, and methane as a function of altitude for the ambient atmosphere.

Figure 2. Pressure-temperature profile for the ambient atmosphere. The temperatures are globally and anually averaged.

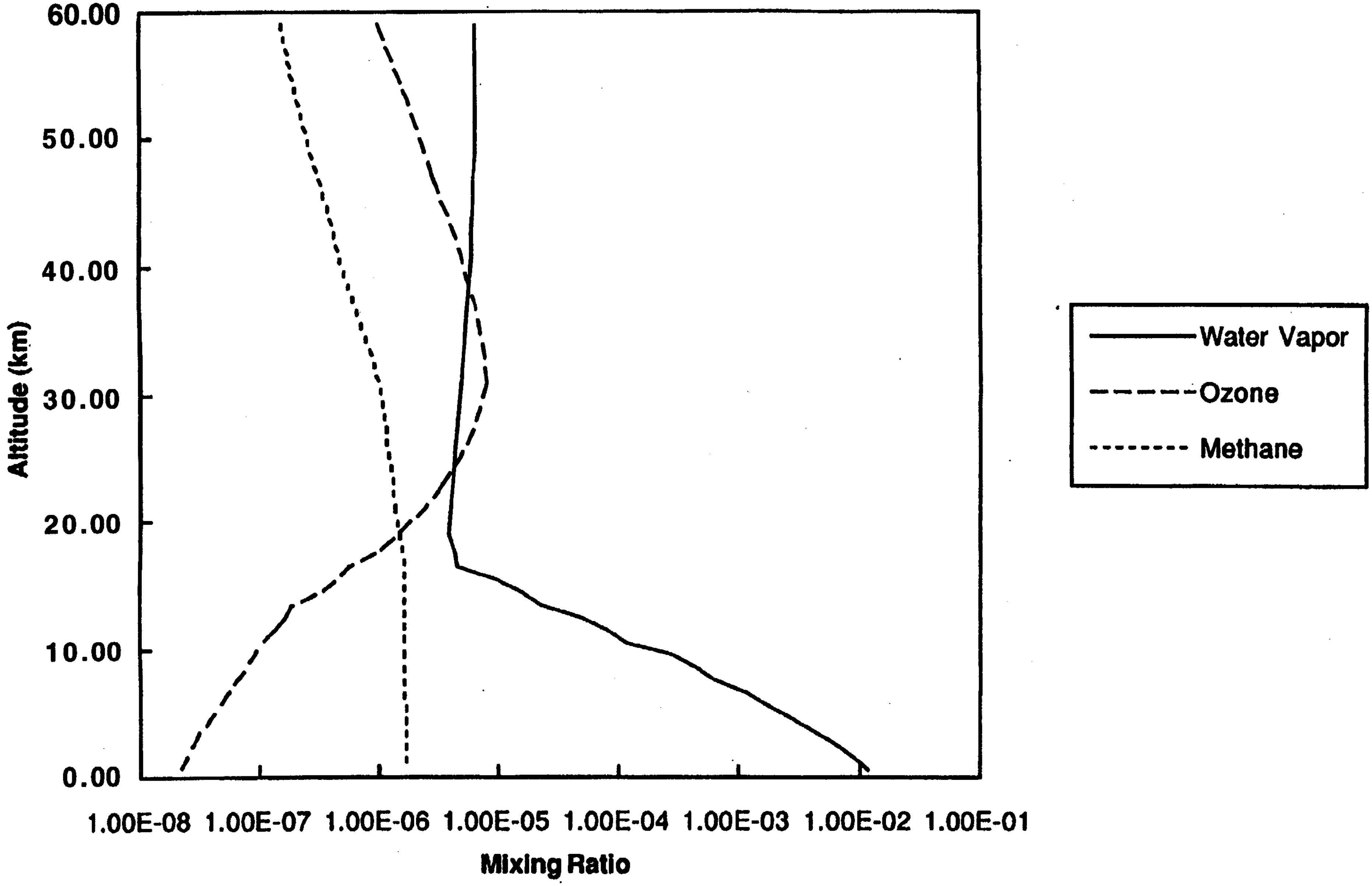

Figure 1 Globally- and annually-averaged profiles of water vapor, ozone, and methane as a function of altitude for the ambient atmosphere.

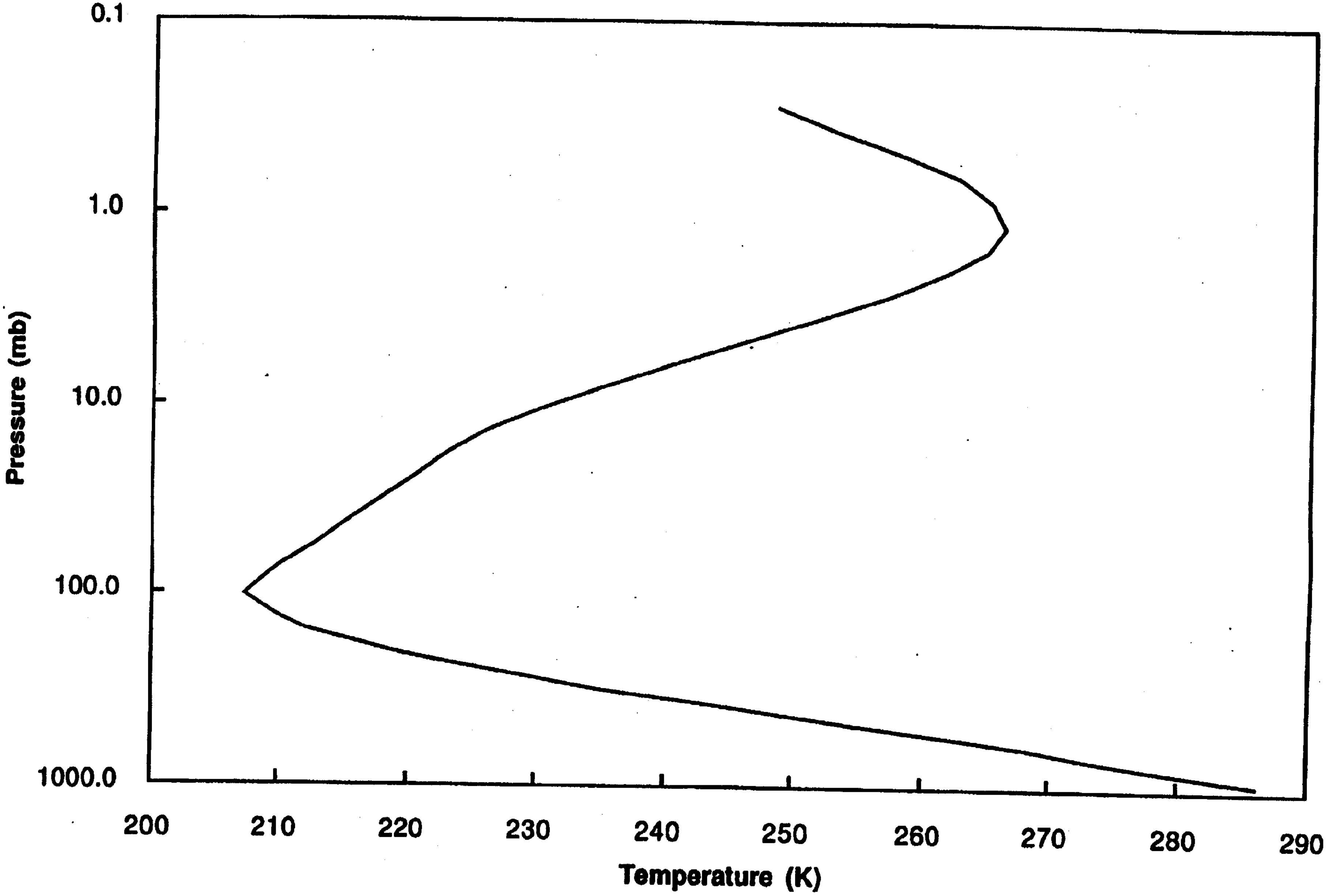

Figure 2 Pressure-temperature profile for ambient atmosphere. The temperatures are globally- and annually-averaged.

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