

## Methane and Nitrous Oxide: Their Effects on the Terrestrial Climate

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### ABSTRACT

Simplified band models are developed for methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) bands in the longwave radiation spectrum. The band models are then employed in a radiation model to calculate the seasonally and latitudinally varying contributions of CH<sub>4</sub> and N<sub>2</sub>O to the radiative energy balance of the earth-troposphere system. From the energy balance calculations, it is concluded that the longwave opacity (i.e., the so-called "greenhouse effect") due to present-day observed concentrations of CH<sub>4</sub> and N<sub>2</sub>O contribute nearly 2 K to hemispherical mean surface temperature with possible larger contributions to polar surface temperatures. The paper also discusses stratospheric effects of CH<sub>4</sub> and N<sub>2</sub>O and examines the sensitivity of tropospheric radiation energy balance to large increases in CH<sub>4</sub> and N<sub>2</sub>O.

### 1. Introduction

Methane and N<sub>2</sub>O possess several strong absorption bands in the longwave radiation spectrum. The strength of these bands, when considered in conjunction with the observed present-day concentrations of CH<sub>4</sub> and N<sub>2</sub>O, suggest that these species may exert a non-negligible influence on the present-day climate. The purpose of the present paper is to examine in detail the effects of these species on the zonal and seasonal radiative energy budget. This problem has not been considered before. However, Ellingson and Gille (1978) have included CH<sub>4</sub> and N<sub>2</sub>O in their radiation model and Wang *et al.* (1976) have estimated with a radiative-convective model the sensitivity of global mean surface temperature to large increases in CH<sub>4</sub> and N<sub>2</sub>O concentrations.

First, we present computationally efficient but accurate band models for longwave bands of CH<sub>4</sub> and N<sub>2</sub>O. Next these band models are employed in a radiation model to estimate the following: effects on the seasonal and latitudinal radiation energy balance of the earth-troposphere system; effects on stratospheric temperatures; and sensitivity of earth-troposphere radiation energy balance to large increases in CH<sub>4</sub> and N<sub>2</sub>O.

### 2. Model details

#### a. Band models

We adopt the band model proposed originally by Cess and Ramanathan (1972) and subsequently

modified slightly by Ramanathan (1976). The model describes the total band absorbance  $A$  as

$$A(U, \beta) = 2A_0 \ln \left\{ 1 + \frac{U}{[4 + U(1 + 1/\beta)]^{1/2}} \right\}, \quad (1)$$

where

$$U = SW/A_0, \quad (2)$$

$$\beta = \beta_0(P/P_0). \quad (3)$$

In Eq. (1),  $U$  is the dimensionless optical depth parameter,  $\beta$  the dimensionless line shape parameter with  $\beta_0$  the value of  $\beta$  at a reference pressure of  $P_0 = 1$  atm where  $P$  is the broadening pressure, and  $A_0(\text{cm}^{-1})$  is the bandwidth parameter. The quantities  $S$  and  $W$  are, respectively, the band intensity [ $\text{cm}^{-1}(\text{cm atm})^{-1}$ ] and the absorber amount ( $\text{cm atm}$ ).

The present analysis includes the 1306  $\text{cm}^{-1}$  band of CH<sub>4</sub> and the 589 and 1285  $\text{cm}^{-1}$  bands of N<sub>2</sub>O. Both these species have several other bands between 2000 and 6000  $\text{cm}^{-1}$  (Cess and Chen, 1975; Kunitomo *et al.*, 1977) whose contributions to terrestrial climate are negligible. For N<sub>2</sub>O absorption in the 1285  $\text{cm}^{-1}$  spectral region, the analysis includes the fundamental and the first hot band and, furthermore, accounts for contribution from four isotopes. These N<sub>2</sub>O bands overlap each other and the extension of (1) to overlapping bands is straightforward, as shown in Ramanathan (1976). The band intensity for the 1306  $\text{cm}^{-1}$  methane band is taken from Cess and Chen, while the N<sub>2</sub>O band intensities are obtained from McClatchey *et al.* (1973). The band parameters  $A_0$  and  $\beta_0$  are obtained as follows. For CH<sub>4</sub>,  $A_0$  and  $\beta_0$  were obtained by fitting  $A$  predicted by (1) to laboratory measurements of  $A$  by

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TABLE 1. Methane and nitrous oxide band model parameters.

Parameter	CH <sub>4</sub> (1306 cm <sup>-1</sup> )	N <sub>2</sub> O (1285 cm <sup>-1</sup> )	N <sub>2</sub> O (589 cm <sup>-1</sup> )
$A_0$ (cm <sup>-1</sup> )	52 (T/300) <sup>1/2</sup>	20.4 (T/300) <sup>1/2</sup>	23 (T/300) <sup>1/2</sup>
$\beta_0$	0.17 (300/T) <sup>1/2</sup>	1.12 (300/T) <sup>1/2</sup>	1.08 (300/T) <sup>1/2</sup>

Burch and Williams (1962a). For the N<sub>2</sub>O band in the 1285 cm<sup>-1</sup> region,  $A$  was calculated from spectral line parameters measured by Kunitomo *et al.* (1977). These line parameters were employed in a statistical spectral band model (Malkmus, 1967) to obtain  $A$  for various values of  $W$  and  $P$ . The computed values of  $A$  were employed in (1) to obtain  $A_0$  and  $\beta_0$ . For the 589 cm<sup>-1</sup> N<sub>2</sub>O band,  $A_0$  and  $\beta_0$  were taken from Kunitomo *et al.* The computed  $A_0$  and  $\beta_0$  values for the various bands are shown in Table 1.

Fig. 1 compares the CH<sub>4</sub> band absorptance computed by (1) with Burch and Williams (1962a) data. We also computed  $A$  by employing the spectral parameters given by Green *et al.* (1964) in the statistical band model. The band was divided into six spectral intervals. The results obtained from these detailed calculations are also shown in Fig. 1. In addition, Table 2 compares values of  $A$  obtained from (1) with those measured by Gryvnak *et al.* (1976). Generally, the agreement between (1) and measured absorptance is as good as the agreement between Green *et al.*'s detailed spectral model results and measurements. Furthermore, Green *et al.*'s model applies only in the strong line limit which is generally not valid for  $W < 0.1$  cm atm. This limitation of Green *et al.*'s model is the source

for the large discrepancy between Eq. (1) and Green *et al.*'s model at small values of  $W$  (see Fig. 1,  $P \approx 0.5$  atm).

Fig. 2 compares the N<sub>2</sub>O band absorptance for the 1285 cm<sup>-1</sup> band as computed by (1) with the detailed band absorption calculations mentioned earlier and with laboratory data of Burch *et al.* (1960). Agreements similar to those shown in Fig. 2 are obtained when comparing (1) with absorptances measured by Burch and Williams (1962b) and by Burch *et al.* (1971).

### b. Radiation model

The radiative energy budget calculations were performed with the radiation model of Ramanathan and Dickinson (1979), modified to include CH<sub>4</sub> and N<sub>2</sub>O. The vertical, latitudinal and seasonal distribution of observed pressure, temperature, humidity, ozone and cloud cover as described in Ramanathan and Dickinson are combined with the CH<sub>4</sub> distribution of the U.S. Standard Atmosphere (1976) and the N<sub>2</sub>O distribution of Crutzen *et al.* (1978) to define an unperturbed or base state atmosphere (surface concentration of CH<sub>4</sub> is 1.5 ppmv; surface concentration of N<sub>2</sub>O is 0.30 ppmv). These observed distributions correspond to the Northern Hemisphere and, hence, our energy budget calculations apply to the Northern Hemisphere. Possible latitudinal variations in the vertical profiles of CH<sub>4</sub> and N<sub>2</sub>O are not considered. The overlap of water vapor bands in the 1306 and 1285 cm<sup>-1</sup> regions with the corresponding CH<sub>4</sub> and N<sub>2</sub>O bands is accounted for by multiplying the band absorptances of CH<sub>4</sub> and N<sub>2</sub>O with the mean transmissivity of water vapor in the region of overlap. Similar

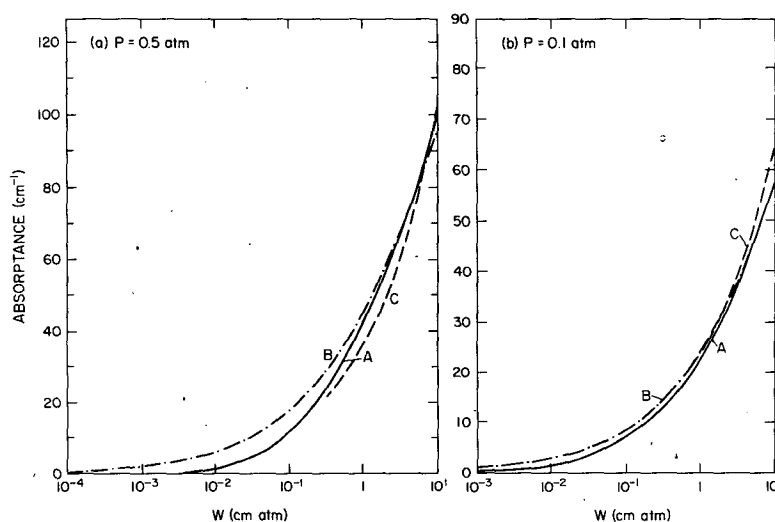


FIG. 1. Comparison of CH<sub>4</sub> 1306 cm<sup>-1</sup> band absorptance. A, Eq. (1) in text; B, Green *et al.*'s (1964) model; C, Burch and Williams' (1962a) data.  $W$  is the CH<sub>4</sub> amount (cm atm) and  $P$  is total pressure.

TABLE 2. Comparison of CH<sub>4</sub> 1306 cm<sup>-1</sup> band absorbance.

Total pressure (atm)	Absorber amount STP (atm cm)	Absorbance (cm <sup>-1</sup> )		
		(1)*	(2)*	Eq. (1)
1.0	0.916	50.1	54.0	51.6
1.0	0.505	35.2	47.8	41.6
1.0	0.458	34.0	41.6	36.5
1.0	0.231	22.8	31.8	24.4
0.3	0.275	18.6	20.9	19.1
0.3	0.137	12.3	15.4	12.8
0.3	0.069	7.2	11.3	8.2
0.1	0.092	7.4	7.9	6.9
0.1	0.046	3.6	5.6	4.5
0.1	0.023	2.5	4.0	2.8
0.03	0.028	2.3	2.4	2.1

\* (1): Gryvnak *et al.*'s (1976) data; (2): calculated from Green *et al.*'s (1964) band model.

treatments were used to take into account overlap of CH<sub>4</sub> and N<sub>2</sub>O bands in the 1285 cm<sup>-1</sup> region and the overlap by N<sub>2</sub>O 589 cm<sup>-1</sup> band with water vapor and carbon dioxide bands. Transmissivities for water vapor were calculated using the formulations of Rodgers and Walshaw (1966), excepting the H<sub>2</sub>O 8–12 μm continuum transmissivity, which was calculated using the method of Roberts *et al.* (1976). Green *et al.*'s (1964) model was used to calculate CH<sub>4</sub> transmissivity in the CH<sub>4</sub>-N<sub>2</sub>O overlap region. The CO<sub>2</sub> transmissivity in the CO<sub>2</sub>-N<sub>2</sub>O overlap region centered at 589 cm<sup>-1</sup> is calculated following the procedure described in Ramanathan (1976, Appendix A).

Various perturbations (adding or removing CH<sub>4</sub> or N<sub>2</sub>O) were introduced after radiative flux calculations were performed for the base state. The tropospheric temperature profile was held fixed. The departure of stratospheric temperatures from the base state profiles were then calculated using the no-dynamic feedback assumption of Ramanathan and Dickinson (1979), in which the stratospheric temperatures are adjusted until the perturbation radiative heating within the stratosphere vanishes.

### 3. Results

The effect of removing CH<sub>4</sub> on the seasonal radiative energy input to the earth-troposphere system ( $\Delta I^T$ ) is shown in Fig. 3a and the annual mean values are shown in the lower half of Fig. 3b. Fig. 3 indicates that removing CH<sub>4</sub> cools the earth-troposphere system by 1–2 W m<sup>-2</sup>, with the strongest effect at low latitudes, and the most pronounced latitudinal variation is during the winter. These latitudinal and seasonal variations are directly caused by corresponding variations in tropospheric temperature profiles. Values of annual mean  $\Delta I^T$  for doubling and quadrupling CH<sub>4</sub> are shown in the upper half of Fig. 3b. The magnitude of  $\Delta I^T$  is less for doubling CH<sub>4</sub> than for removing CH<sub>4</sub> entirely, a consequence of increasing saturation of the line centers as the concentration of CH<sub>4</sub> increases. The effects of perturbing N<sub>2</sub>O concentration are shown in Fig. 4. The magnitude and latitudinal and seasonal variations of  $\Delta I^T$  are quite similar to those for CH<sub>4</sub>.

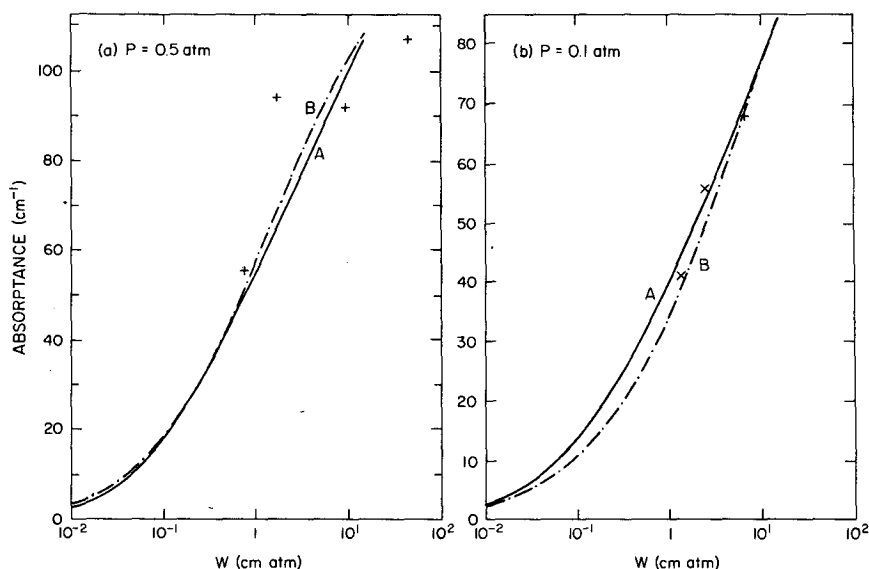


FIG. 2. As in Figure 1 except for 1285 cm<sup>-1</sup> N<sub>2</sub>O bands. A, Eq. 1; B, statistical band model calculations from Kunitomo *et al.*'s (1977) line parameters. Laboratory data from Burch *et al.* (1960) is also shown by '+'s and 'x's, the former denoting  $P$  10–15% greater than that shown in the figure and the latter  $P$  within  $\pm 5\%$  of the value shown.

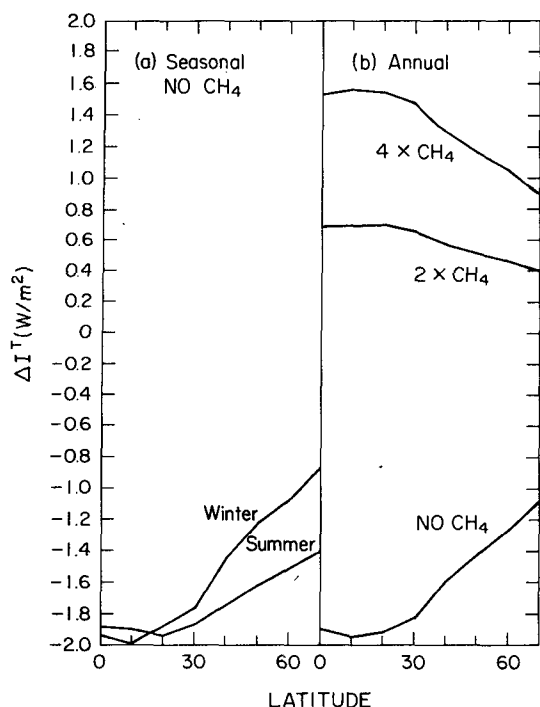


FIG. 3. Effect of  $\text{CH}_4$  on the earth-troposphere radiative energy input.  $\Delta I^T$  is the change in the energy input. (a) Seasonal values for removal of  $\text{CH}_4$ ; (b) annual mean values for removal of  $\text{CH}_4$  (the NO  $\text{CH}_4$  curve), for doubling and quadrupling of  $\text{CH}_4$ .

The effect of removing  $\text{CH}_4$  or  $\text{N}_2\text{O}$  on the zonally and annually averaged stratospheric temperature profile at  $40^\circ\text{N}$  is illustrated in Fig. 5. Again, the no-dynamic feedback scheme of Ramanathan and Dickinson (1979) has been employed. Cooling in the lower stratosphere and heating further aloft is shown in Fig. 5. The temperature change profile has latitudinal variations, with the greatest temperature changes occurring at low latitudes (not shown in the figure). The cooling resulting from combined removal of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  is  $\sim 0.8$  K in winter at the equator in the lower stratosphere. The role of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  in the hemispherical annual mean energy balance of the stratosphere is given in Table 3. These estimates were made by adding  $\text{CH}_4$  and  $\text{N}_2\text{O}$  effects in the model, but holding the stratospheric temperatures fixed at the observed values.

The hemispherical annual mean contributions to the earth-troposphere radiative heating from  $\text{CH}_4$  and  $\text{N}_2\text{O}$  are  $1.7$  and  $1.4$   $\text{W m}^{-2}$  respectively, (obtained from Figs. 3b and 4b, respectively). We can relate the change in hemispherical annual mean earth-troposphere radiative heating  $\langle \Delta I^T \rangle$  to a corresponding change in surface temperature  $\langle \Delta T_s \rangle$  by

$$\langle \Delta T_s \rangle \approx 0.8 \langle \Delta I^T \rangle, \quad (4)$$

where the coefficient  $0.8$   $\text{W m}^{-2} \text{ K}^{-1}$  has been obtained from Wetherald and Manabe's (1975) GCM

experiments which evaluated the sensitivity of the GCM climate to a change in solar constant. The procedure for obtaining this coefficient from the GCM experiments is described in Ramanathan (1977). The  $\langle \Delta I^T \rangle$  value for  $\text{CH}_4$  and  $\text{N}_2\text{O}$  is  $3.1$   $\text{W m}^{-2}$  and from (4) it is seen that these two species may contribute about  $2$  K warming to the hemispherical mean surface temperature with significantly larger (by a factor of 2–3) contributions to polar regions due to ice-albedo feedback (cf., Wetherald and Manabe, 1975).

Wang *et al.* (1976) provide radiative-convective model estimates for the change in surface temperature due to a doubling of  $\text{CH}_4$  and  $\text{N}_2\text{O}$ . A direct comparison between Wang *et al.*'s results and the present results is precluded because Wang *et al.* do not give results for  $\Delta I^T$ . Furthermore, Wang *et al.* perform one-dimensional model calculations, whereas the present analysis performs latitudinal and seasonal calculations to obtain hemispherical mean  $\Delta I^T$ . In spite of these differences, we provide below an approximate estimate of the differences between the two model results. As discussed in Ramanathan and Coakley (1978), the relation between  $\langle \Delta I^T \rangle$  and  $\langle \Delta T_s \rangle$  for a radiative-convective model with fixed relative humidity and fixed cloud altitude is given by

$$\langle \Delta T_s \rangle \approx 0.5 \langle \Delta I^T \rangle. \quad (5)$$

The coefficient  $0.5$  in (5) is smaller than the  $0.8$  value shown in (4) which applies to GCM results.

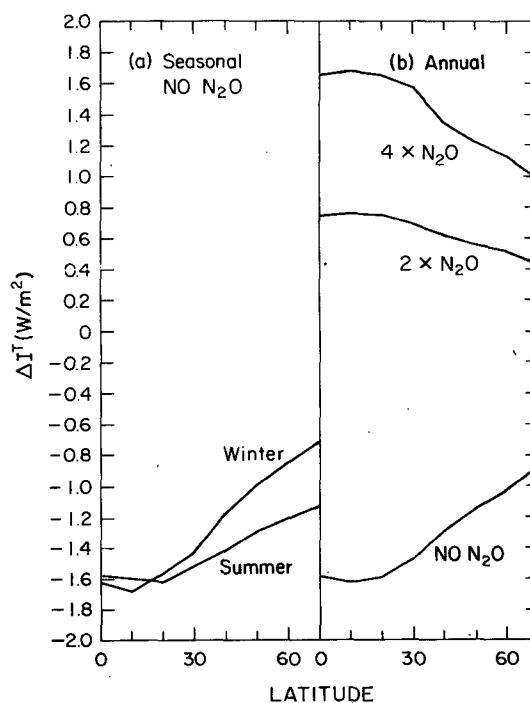


FIG. 4. As in Fig. 3 except for  $\text{N}_2\text{O}$ .

The GCM value is larger mostly due to the inclusion of ice-albedo feedback which is neglected in the radiative-convective model. From Figs. 3b and 4b, the hemispherical mean  $\Delta I^T$  for doubled  $\text{CH}_4$  is  $0.61 \text{ W m}^{-2}$  and that for  $\text{N}_2\text{O}$  is  $0.65 \text{ W m}^{-2}$ . From (5),  $\langle \Delta T_s \rangle \approx 0.3 \text{ K}$  for both doubled  $\text{CH}_4$  and for doubled  $\text{N}_2\text{O}$ . Wang *et al.* (1976) estimate 0.2 K and 0.44 K, respectively, for doubled  $\text{CH}_4$  and doubled  $\text{N}_2\text{O}$ .

The present estimate of  $\langle \Delta T_s \rangle$  for  $\text{CH}_4$  doubling is somewhat larger than that of Wang *et al.* However, Wang *et al.* obtained  $\langle \Delta T_s \rangle \approx 0.4 \text{ K}$  for  $\text{CH}_4$  doubling by using a different set of absorption data. The value of  $\langle \Delta T_s \rangle$  estimated in the present analysis lies between the low and high values computed by Wang *et al.* The present estimate of  $\langle \Delta T_s \rangle$  for doubling  $\text{N}_2\text{O}$  is smaller than that of Wang *et al.* by about 30%. Wang *et al.* include the  $\text{N}_2\text{O}$  band centered at  $2224 \text{ cm}^{-1}$ . This band is neglected in the present analysis since its contribution to  $\langle \Delta T_s \rangle$  is negligible. The 30% difference in the  $\langle \Delta T_s \rangle$  computed by the two models (for doubling of  $\text{N}_2\text{O}$ ) is perhaps due to the following modeling differences: (i) Wang *et al.*'s is a one-dimensional model calculation as opposed to the hemispherical averaging procedure employed in the present model; and (ii) the two models treat  $\text{CO}_2$  overlap effects differ-

TABLE 3. Contribution of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  to the annual mean radiative energy balance of the Northern Hemisphere stratosphere. All units  $\text{W m}^{-2}$ . NET = absorption - emission.

	$\text{CH}_4$	$\text{N}_2\text{O}$
Emission	0.30	0.41
Absorption	0.27	0.45
NET	-0.03	0.04

ently in the  $589 \text{ cm}^{-1}$  region. The transmissivity of  $\text{CO}_2$  in this region is a rapidly varying function of wavenumber, which makes it difficult to define a mean  $\text{CO}_2$  transmissivity for the entire  $\text{N}_2\text{O}$  band. Consequently there could be considerable differences between the two models in the computed mean  $\text{CO}_2$  transmissivity. This possible difference may contribute appreciably to the difference in  $\langle \Delta T_s \rangle$  between the two models, particularly in view of the fact that  $\Delta I^T$  due to  $\text{N}_2\text{O}$  is strongly dependent on  $\text{CO}_2$  overlap effects (e.g., for  $\text{N}_2\text{O}$  doubling,  $\Delta I^T$  without  $\text{CO}_2$  overlap is about 60% larger than that with  $\text{CO}_2$  overlap).

## 5. Summary

The latitudinally and seasonally varying contributions of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  (in their present-day concentrations) to the radiative energy budget of the surface-troposphere system are large enough to contribute approximately 2 K to the hemispherical mean surface temperature with possibly larger contributions to polar temperatures. The present results indicate the necessity for including the radiative effects of  $\text{CH}_4$  and  $\text{N}_2\text{O}$  in climate models, at least in those climate models which explicitly allow for the radiative effects of various optically active atmospheric constituents.

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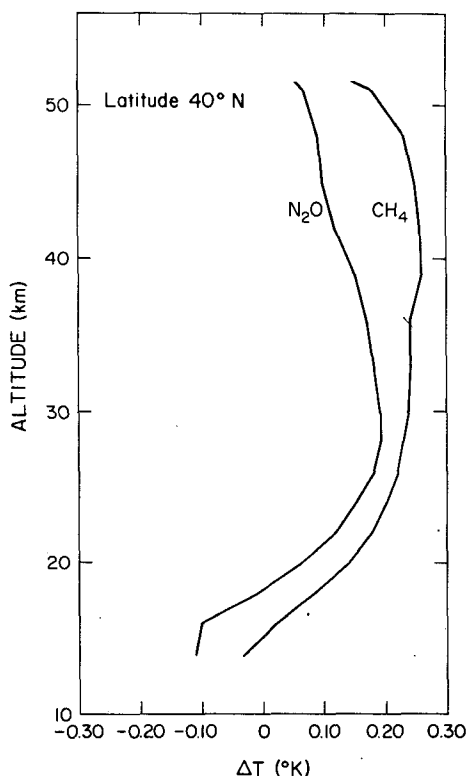


FIG. 5. Annual mean effect of removing  $\text{CH}_4$  and  $\text{N}_2\text{O}$  at  $40^\circ\text{N}$  on stratospheric temperatures.  $\Delta T$  is the change in stratospheric temperature from the observed values.

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