

Infrared radiative forcing and atmospheric lifetimes of trace species based on observations from UARS

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Abstract. Observations from instruments on the Upper Atmosphere Research Satellite (UARS) have been used to constrain calculations of infrared radiative forcing by CH₄, CCl₂F₂, and N₂O and to determine lifetimes of CCl₂F₂ and N₂O. Radiative forcing is calculated as a change in net infrared flux at the tropopause that results from an increase in trace gas amount from preindustrial (1750) to contemporary (1992) times. Latitudinal and seasonal variations are considered explicitly, using distributions of trace gases and temperature in the stratosphere from UARS measurements and seasonally averaged cloud statistics from the International Satellite Cloud Climatology Project. Top-of-atmosphere fluxes calculated for the contemporary period are in good agreement with satellite measurements from the Earth Radiation Budget Experiment. Globally averaged values of the radiative forcing are 0.550, 0.132, and 0.111 W m⁻² for CH₄, CCl₂F₂, and N₂O, respectively. The largest forcing occurs near subtropical latitudes during summer, predominantly as a result of the combination of cloud-free skies and a high, cold tropopause. Clouds are found to play a significant role in regulating infrared forcing, reducing the magnitude of the forcing by 30–40% compared with the case of clear skies. The vertical profile of CCl₂F₂ is important in determining its radiative forcing; use of a height-independent mixing ratio in the stratosphere leads to an overprediction of the forcing by 10%. The impact of stratospheric profiles on radiative forcing by CH₄ and N₂O is 2% or less. UARS-based distributions of CCl₂F₂ and N₂O are used also to determine global destruction rates and instantaneous lifetimes of these gases. Rates of photolytic destruction in the stratosphere are calculated using solar ultraviolet irradiances measured on UARS and a line-by-line model of absorption in the oxygen Schumann-Runge bands. Lifetimes are 114 ± 22 and 118 ± 25 years for CCl₂F₂ and N₂O, respectively.

1. Introduction

Increases in the atmospheric abundances of CH₄, CCl₂F₂ (CFC-12) and N₂O from the preindustrial to the present time have had a significant effect on the radiative and photochemical state of the atmosphere.

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The enhanced infrared opacity as a result of these changes accounts for nearly one third of the postindustrial, greenhouse gas direct radiative forcing [Intergovernmental Panel on Climate Change (IPCC), 1995]. In addition, CH₄, CCl₂F₂, and N₂O are significant sources of hydrogen, chlorine, and nitrogen radicals in the stratosphere and thus play an important role in the budget of stratospheric ozone. Global destruction rates of CCl₂F₂ and N₂O are determined primarily by ultraviolet photolysis in the stratosphere.

Although the longwave effects of trace gas increases were recognized some time ago [Ramanathan, 1975; Wang *et al.*, 1976], it is only recently that global distributions have become available for including latitudinal

as well as seasonal effects. Calculations of radiative forcing, for the most part, have been based on results using global mean vertical profiles of temperature, water vapor, and ozone [Wang *et al.*, 1980; Hansen *et al.*, 1981; Ramanathan *et al.*, 1987]. The computed flux, however, may not be identical to the global average of separate calculations at each latitude [see, for example, *World Meteorological Organization (WMO)*, Chapter 7 1991; Pinnock *et al.*, 1995; Myhre and Stordal, 1997; Freckleton *et al.*, 1998]. Furthermore, the decrease in concentrations of CH₄, CCl₂F₂, and N₂O with increasing altitude above the tropopause can influence the downward flux at the tropopause level, thereby impacting the radiative forcing [Ramanathan *et al.*, 1985]. These effects are accounted for implicitly within the context of two- and three-dimensional model studies [Kiehl and Briegleb, 1993; Hauglustaine *et al.*, 1994; Hansen *et al.*, 1997], with trace gas distributions determined internally by the model.

The importance of CH₄, N₂O, and most CFCs to the Earth's longwave radiation budget is due largely to the spectral location of the principal IR bands involved. Most are located within, or near, a relatively transparent spectral window between 8 and 12 μm . The accurate specification of stratospheric ozone in the radiative calculations is thus important due to the presence of the O₃ 9.6- μm band. Equally important, tropospheric clouds are strong sources of opacity in the 8 to 12- μm window, and the proper representation of clouds is critical to the evaluation of radiative forcing at the tropopause. High clouds in the troposphere have been shown to influence strongly the stratospheric IR heating by ozone [e.g., Dessler *et al.*, 1996], as well as the radiative forcing by tropospheric O₃ [Forster *et al.*, 1996] and CFC replacements [Pinnock *et al.*, 1995].

The destruction of N₂O and CCl₂F₂ is thought to take place primarily in the stratosphere. Most of the loss occurs through photodissociation by absorption of solar ultraviolet radiation in the 190- to 220-nm spectral region, at altitudes between 20 and 40 km [Minschwaner *et al.*, 1993]. Lifetimes for N₂O and CFC compounds have been estimated using two-dimensional photochemical models of the middle atmosphere [e.g., Ko and Sze, 1982], with calculated values determined by the balance between stratospheric loss and influx to the stratosphere by cross-tropopause transport in the tropics. A more empirical method applies calculated destruction rates to observed distributions in the stratosphere [Johnston *et al.*, 1979]. If the source terms are accurately known, then the lifetime can also be estimated from the total atmospheric burden [e.g., Kaye *et al.*, 1994]. A novel technique outlined by Plumb and Ko [1992] employs correlations between long-lived tracers in the stratosphere to determine lifetime ratios. The use of this technique is discussed within the framework of three-dimensional model calculations by Avallone and Prather [1997] and is applied to ER-2 observations in the stratosphere by Volk *et al.* [1997].

The present study is motivated by the availability of global-scale observations from the Upper Atmosphere Research Satellite (UARS), which are relevant to the

evaluation of radiative forcings and lifetimes of trace gases. UARS measurements of CH₄, CCl₂F₂, N₂O, O₃, H₂O, temperature, and solar ultraviolet irradiances are used to represent the physical and chemical state of the contemporary stratosphere. Complementary measurements of trace gas mixing ratios at the surface, mean cloud statistics from the International Satellite Cloud Climatology Project (ISCCP), and average longwave fluxes from the Earth Radiation Budget Experiment (ERBE) are used to complete this picture. The goal is to take advantage of the unprecedented spatial and temporal coverage of UARS measurements and apply them to the evaluation of radiative forcing by CH₄, CCl₂F₂, and N₂O and to determine atmospheric lifetimes for CCl₂F₂ and N₂O.

2. UARS Distributions of Trace Gases and Temperature

Climatological distributions of trace gases were constructed from zonal averages of UARS observations over 2-month periods bracketing the equinoxes and solstices. The total time interval extends from March 1992 to January 1993. All calculations were carried out on a 5° latitude grid from 77.5°S to 77.5°N. Constant values were used poleward of 77.5° for the purposes of obtaining global averages. Vertical levels were spaced 100 mbar apart in the troposphere, decreasing to 10-20 mbar near the tropopause and lower stratosphere. Vertical resolution in the middle and upper stratosphere was approximately 3 km.

Distributions of CH₄, CCl₂F₂, and N₂O in the stratosphere were derived from vertical profiles measured by the Cryogen Limb Array Etalon Spectrometer (CLAES) on board UARS. The CLAES instrument observed limb emission in vibration-rotation spectra between 3.5 and 13 μm for retrieval of trace gas mixing ratios, pressure, and temperature [Roche *et al.*, 1993]. Version 7, level 3AT data were used for all three gases. Averages for each latitude bin were obtained by weighting all values according to the data quality indicator associated with each point. The zonally averaged vertical profile was linearly interpolated from the UARS standard pressure levels to our adopted vertical grid. CLAES data were used from 0.1 mbar down to 46 mbar; values below this level were determined using a cubic spline interpolation to the tropopause (specification of the tropopause pressure is discussed below).

Mixing ratios in the troposphere, assumed to be height independent due to mixing, were obtained from 1991-1992 measurements from the surface network operated by the Climate Monitoring and Diagnostics Laboratory for CH₄ [Tans *et al.*, 1992] and for CCl₂F₂ and N₂O [Montzka *et al.*, 1992]. We approximated latitudinal gradients in mixing ratios using linear fits to data from six surface stations ranging from 82.5°N to 40.7°S. This variation is small for N₂O (less than 1% from pole to pole) but is significant for CH₄ (~9% pole to pole) and CCl₂F₂ (~6%).

Other model inputs which are constrained by data from UARS include stratospheric O₃ and H₂O. Cor-

rect specification of ozone is of some importance for the infrared flux due to spectral overlaps in the wings of the O₃ 9.6- μ m band and the N₂O 7.8- μ m band, the CH₄ 7.7- μ m band, and CCl₂F₂ absorption bands near 9 μ m. The 6.7- μ m band of water has an effect also on the N₂O and CH₄ bands. In the ultraviolet, the short-wavelength tail of the ozone Hartley band contributes significantly to the opacity near 200 nm and therefore impacts photodissociation of both CCl₂F₂ and N₂O. Zonally averaged distributions of O₃ and H₂O were compiled based on measurements from the UARS Microwave Limb Sounder (MLS), which observes limb emission at millimeter wavelengths [Waters 1993; Froidevaux *et al.*, 1994]. Version 3, level 3AT data from the 205-GHz radiometer were used for O₃; data from the 183-GHz radiometer were used for H₂O. Zonal and seasonal averages of MLS measurements above 50 mbar were constructed in a manner similar to the CLAES data described above. Ozone in the troposphere was based on climatological values from Oltmans [1981] and Levy *et al.* [1985]. Tropospheric water vapor was specified on the basis of standard Air Force Geophysics Laboratory (AFGL) models [Anderson *et al.*, 1986] with values interpolated in season and latitude.

The vertical profile of temperature is another important parameter in the calculation of infrared forcing. Temperatures in the upper atmosphere have an impact also on ultraviolet photolysis through the temperature dependence of absorption cross sections, most notably in the O₂ Schumann-Runge (S-R) bands, with a corresponding effect on the penetration of ultraviolet radiation to the middle and lower stratosphere. Temperatures were adopted from the National Meteorological Center (NMC) analysis [McPherson *et al.*, 1979], which are included as correlative data in the UARS data distribution. NMC temperatures were averaged over season and latitudes, similar to the CLAES trace gas data, from the surface to 70 mbar. CLAES measurements of temperature were used above 70 mbar. These agree well ($\approx \pm 1$ K) with lidar and rocketsonde measurements of temperature up to the 1-mbar pressure level.

As discussed by Myhre and Stordal [1997] and Freckleton *et al.* [1998], specification of the tropopause pressure is a crucial aspect of determining the magnitude of radiative forcing. Owing to changes in tropopause pressure with latitude, adoption of a global-mean tropopause height can produce errors of up to 10% in the instantaneous, global-mean forcing [Myhre and Stordal, 1997]. In addition, Freckleton *et al.* [1998] showed that the choice of tropopause definition (i.e., lapse rate criteria, temperature minimum, top of convective level) can influence radiative forcing results by up to 9%. For the calculations presented below, we defined the tropopause height according to location of minima in temperature profiles at each latitude.

Distributions of CH₄, CCl₂F₂, N₂O, and O₃ for the September-October 1992 time period are shown in Figure 1. The dashed horizontal lines indicate the pressure level which divides UARS-based distributions from the region of interpolation. The tropopause pressure is shown by the bottom dashed curve in each panel. Of

particular importance for our calculations are the decreases in mixing ratios of CH₄, CCl₂F₂, and N₂O in the stratosphere as well as latitudinal variations in vertical profiles. Distributions of H₂O and temperature for the same time period are shown in Figure 2, along with the distributions of cloud fraction and heights assumed in the calculations. These are derived from averages over the 1983-1990 time period for low, middle, and high cloud fractions from the International Satellite Cloud Climatology Project (ISCCP) [Rossow and Schiffer, 1991]. Low clouds are assumed to be located between 850 and 750 mbar, independent of latitude. Middle-level clouds are placed in a 100-mbar-thick layer centered near 500 mbar at middle and high latitudes, and near 550 mbar in tropics, as shown in Figure 2. High clouds are located in a 20-mbar-thick layer located just below the tropopause.

3. Radiative Calculations

As discussed by IPCC [1995], the concept of radiative forcing is a valuable measure of the first-order climatic impact of a greenhouse gas. The forcing is defined by the change in net infrared flux at the tropopause due to a prescribed change in greenhouse gas amount, holding all other model parameters fixed (except for stratospheric temperatures, as discussed below). Adoption of the tropopause as a reference is motivated by the fact that a change in radiative flux at this level appropriately expresses the radiative forcing of the climate system as a whole, since the surface and troposphere are a tightly coupled thermodynamic system. In addition, defining the forcing in terms of the radiative flux change avoids uncertainties associated with a given climate response, for example, surface temperature, which depends on sensitivities and feedbacks that are evidently model-dependent [e.g., IPCC, 1990]. However, care must be taken to account for the temperature response in the stratosphere [Hansen *et al.*, 1981; Ramanathan *et al.*, 1987]. By maintaining radiative equilibrium (or constant heating/cooling) in the stratosphere, any change in flux at the tropopause is then the same as at the top of the atmosphere (TOA).

Infrared fluxes were calculated using a longwave-band model developed at the National Center for Atmospheric Research. This is a 100-cm⁻¹ band model which includes infrared opacity by H₂O, CO₂, and O₃, and considers also the major absorption bands of CH₄, N₂O, CFC-11, and CFC-12. The code is similar in many respects to the Community Climate Model, Version 2 (CCM2) radiation code described by Briegleb [1992]. Line parameters for H₂O, CO₂, O₃, CH₄, and N₂O are based on the HITRAN database [Rothman, 1986]. Absorption cross sections for CFC-11 and CFC-12, averaged over the 100 cm⁻¹ bands, are from Massie *et al.* [1991]. For the trace gases of interest here, the important longwave bands considered include the 7.7- μ m band of CH₄, the 4.5-, 7.8-, and 17- μ m bands of N₂O (see Figure 2 of Briegleb [1992]), and the 8.7-, 9.1-, and 10.9- μ m bands of CCl₂F₂.

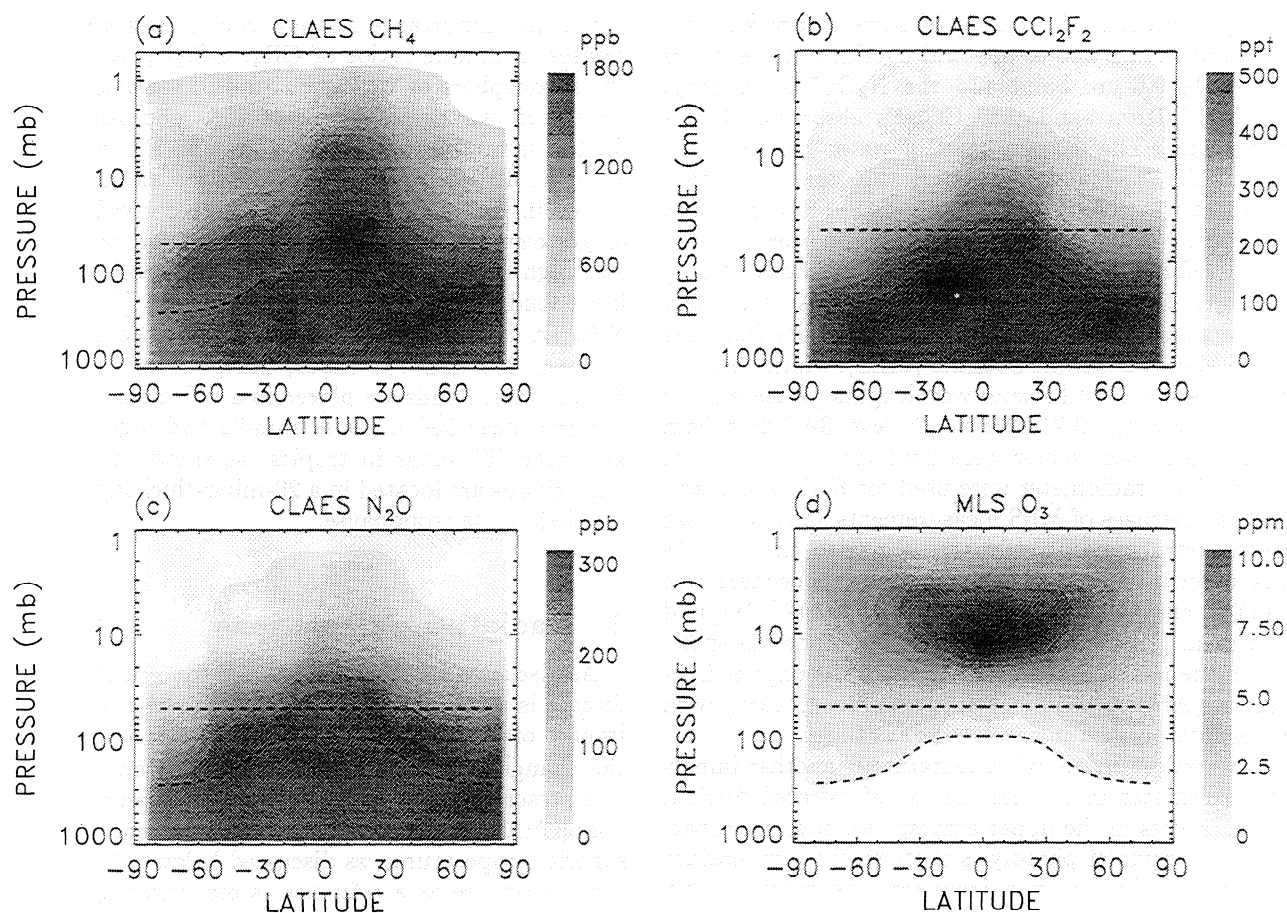


Figure 1. Distributions of (a) CH₄ (in ppb), (b) CCl₂F₂ (in ppt), (c) N₂O (in ppb), and (d) O₃ (in ppm) used in the radiative calculations for the September-October contemporary period. Dashed curves indicate the tropopause and the lowest pressure level for UARS data. Tropospheric mixing ratios are specified as described in the text.

Net longwave fluxes calculated with this model agree with line-by-line calculations to within 1% [Briegleb, 1992]. We have, in addition, compared results for flux changes induced by changes in CH₄ and N₂O with line-by-line calculations by Clough and Iacono [1995]. Results at the tropopause agree to within 10%. Figure 3 compares the TOA flux calculated with the model with measurements from the Earth Radiation Budget Experiment (ERBE) [Barkstrom, 1984] averaged over the 1985-1988 period. The agreement is within the uncertainties in flux ($\pm 5 \text{ W m}^{-2}$) for nearly all latitudes and seasons. Comparison of model shortwave albedo with ERBE averages (not shown) also indicate a good level of agreement.

Results for June-July (Figure 3a) clearly show the impact of high clouds near the Intertropical Convergence Zone between 5° and 10°N which give rise to a minimum in TOA flux. This minimum shifts to south of the equator in both the ERBE observations and model calculations for December-January (Figure 3b). Calculated TOA fluxes are sensitive to assumptions of cloud liquid water paths (*lwp*) and effective drop radii (*r_e*) assumed in the model. We adopted values for the three cloud types that are consistent with measurements [Stephens,

1978; Stephens and Platt, 1987]; these are identical to the values used by Dessler *et al.* [1996] (*lwp* = 125, 75, 15 g m⁻², and *r_e* = 10, 11, 18 μm, for low, middle, and high clouds, respectively). Surface emissivity was fixed at 0.92, independent of latitude and season. This value represents an estimate of the weighted mean of sea surface (*e* ≈ 0.92-0.96), sand (0.85-0.90), and snow (0.85-0.99). Calculated forcings were found to be relatively insensitive to the precise choice of surface longwave emissivity.

For the determination of instantaneous lifetimes, photochemical loss due to ultraviolet photodissociation was calculated using the high-resolution radiation code described by Minschwaner *et al.* [1993]. Solar irradiances above the atmosphere were specified according to measurements from the Solar Stellar Irradiance Comparison Experiment (SOLSTICE) on board UARS, averaged over the month of March 1992. These are high-resolution (0.1 nm) measurements of the full-disk, solar spectral irradiance between 115 and 420 nm [Rottman *et al.*, 1993]. Calibration is maintained to within 1% using a collection of bright blue stars as radiance standards [Woods *et al.*, 1993]. Version 7, level 3BS irradiances with a spectral resolution of 1 nm were used

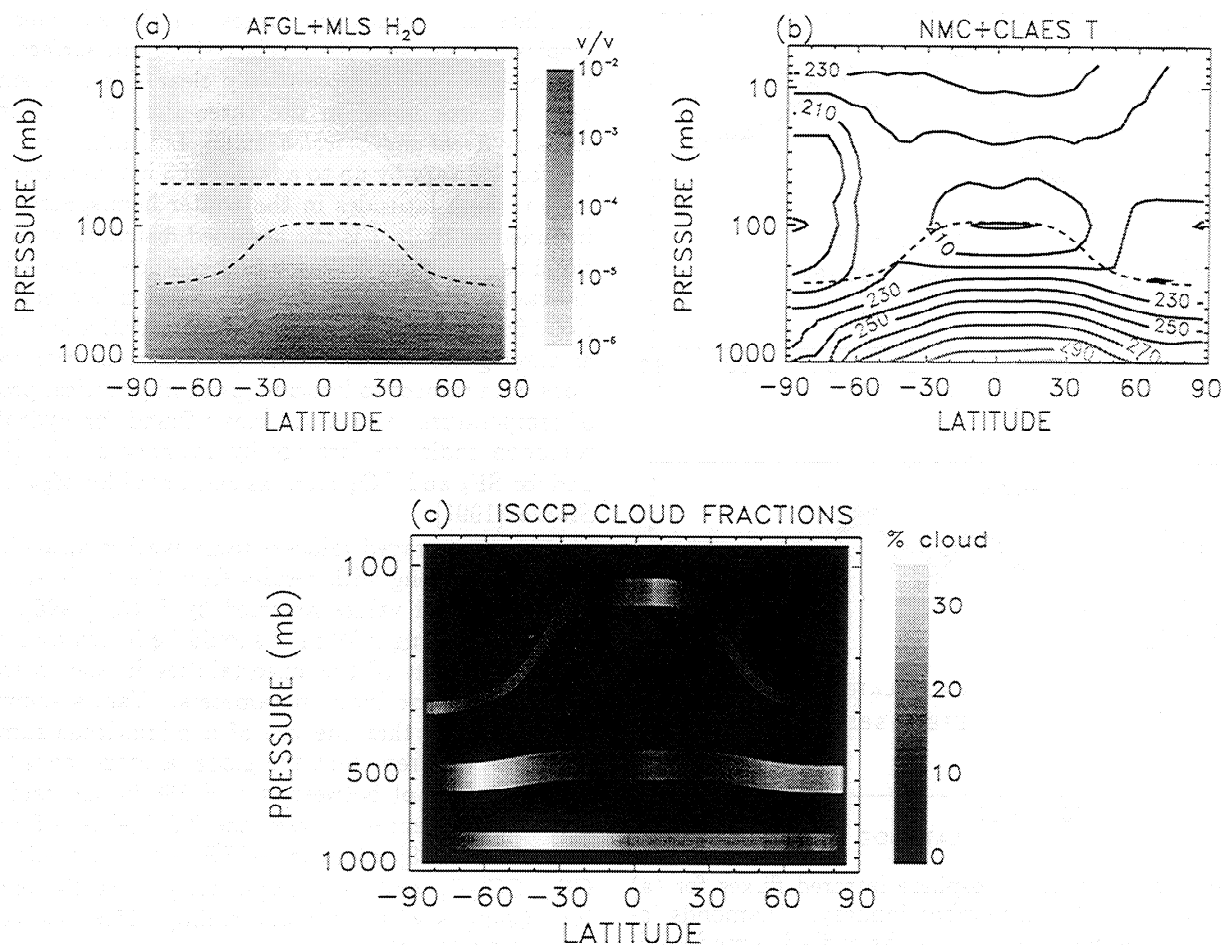


Figure 2. Distributions of (a) H₂O (volume mixing ratio), (b) temperature (degrees Kelvin), and (c) cloud fractions (percent) used in the radiative calculations for the September-October preindustrial and contemporary time periods.

over the full spectral range. Oxygen cross sections in the S-R band region followed the line-by-line analysis by *Minschwaner et al.* [1992]. Cross sections for the O₂ Herzberg continuum were adopted from *Yoshino et al.* [1988], and cross sections for O₃, CCl₂F₂, and N₂O were adopted from *DeMore et al.* [1994]. Below 205 nm, solar irradiances and all cross sections other than O₂ were linearly interpolated to the high-resolution (0.002 nm) spectral grid necessary to capture the rotational structure in O₂ S-R band absorption. Effects of scattering above 190 nm were included based on the formulation of *Meier et al.* [1982].

The instantaneous lifetime for each gas was determined from the ratio of its 1992 global inventory M by the calculated global removal rate L :

$$\tau(1992) = M/L \quad (1)$$

The two quantities in the above equations are defined by

$$M = 2\pi R_E^2 \int dz \int n(\phi, z, 1992) \cos \phi d\phi \quad (2)$$

$$L = 2\pi R_E^2 \int dt \int dz \int n(\phi, z, t) J(\phi, z, t) \cos \phi d\phi \quad (3)$$

where $n(\phi, z, t)$ is the local concentration at latitude ϕ , altitude z , and time t and where R_E is the Earth's radius. The corresponding loss frequency J is set primarily by photodissociation, although total loss includes a small contribution (5-10%) due to reaction with O(¹D). Global fields of O(¹D) were calculated based on a steady state between production due to photolysis of ozone, using the MLS ozone fields described above, and loss due to quenching by N₂ and O₂. Rates for quenching and for reactions of O(¹D) with CCl₂F₂ and N₂O followed recommended values by *DeMore et al.* [1994].

4. Results

4.1. Radiative Forcing

The radiative forcing from preindustrial (1750) to the 1992 time period was calculated using preindustrial mixing ratios of 700 ppb CH₄, 0 ppt CCl₂F₂, and 275 ppb N₂O in the troposphere [*IPCC*, 1995]. Except for CCl₂F₂, there are no corresponding estimates for stratospheric distributions. A reasonable approximation, however, is to assume the same relative distribu-

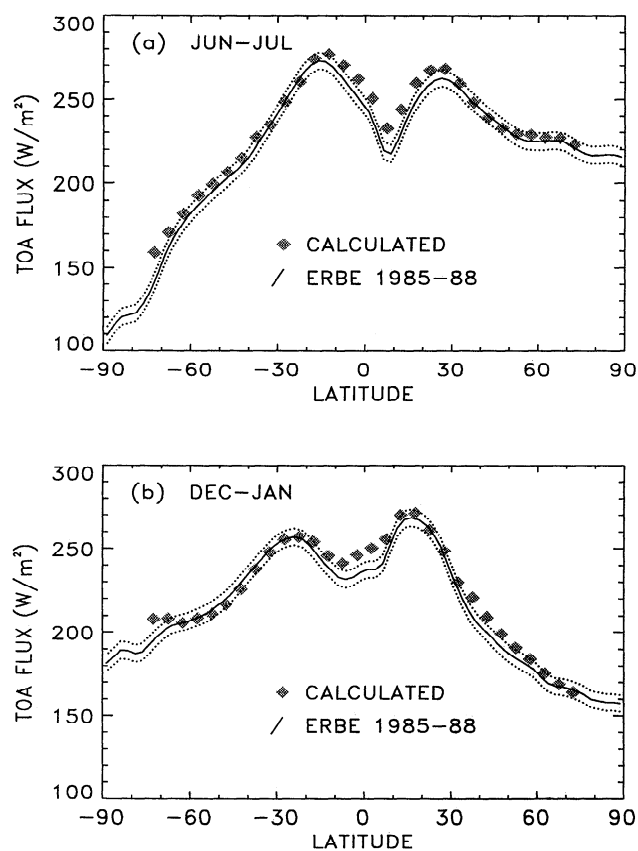


Figure 3. Top of atmosphere infrared fluxes for (a) June-July and (b) December-January. Diamonds indicate values calculated using the radiative model initialized with temperature, trace gas distributions, and clouds as described in the text. Solid curves represent averages of 1985-1988 observations from the Earth Radiation Budget Experiment.

tion as is currently observed. This follows from the fact that the stratospheric loss scales linearly with trace gas amount. Therefore preindustrial trace gas distributions were estimated by scaling all vertical profiles according to the ratio of preindustrial to present-day mixing ratios in the troposphere. It should be noted that the radiative forcing for each gas was computed by using the observed change in abundance of that gas alone, with all other trace gases held fixed at their preindustrial amounts.

The model was modified for the study of radiative forcing by allowing stratospheric temperatures to adjust so that stratospheric heating/cooling rates were identical to values calculated for the preindustrial stratosphere. This approach essentially assumes a fixed dynamical heating/cooling in the stratosphere rather than complete radiative equilibrium, but the final results should be comparable [Ramanathan and Dickinson, 1979]. Temperatures were adjusted iteratively until the net heating rate changed by less than 5×10^{-4} K day $^{-1}$ at all levels above the tropopause.

The magnitude of radiative forcing for CH₄, CCl₂F₂, and N₂O as a function of season and latitude is shown

in Plate 1. Values are largest in the summer subtropics, consistent with a relatively warm surface, cold tropopause, and comparatively clear skies. A similar behavior was noted in the three-dimensional simulations by Kiehl and Briegleb [1993]. Latitudinal values of the forcing vary by up to a factor of 3 between the tropics and high latitudes in the winter hemisphere. Also included in Plate 1 is the assumed distribution of high cloud as a function of season and latitude. The patterns indicate a high degree of anticorrelation between high cloud fraction and the magnitude of radiative forcing for all three gases. These results are consistent with variations with respect to latitude (primarily vertical profiles of temperature) and cloud cover found for hydrofluorocarbon radiative forcings by Pinnock *et al.* [1995] and for SF₆ and CO₂ forcings calculated by Myhre and Stordal [1997].

Globally averaged values of radiative forcing are listed in Table 1, along with results from the 42°N summer calculation and values reported by IPCC [1995]. The digit representing mW m $^{-2}$ should be interpreted with caution in light of the uncertainties in spectroscopic and atmospheric input parameters. Values shown in Table 1 imply that the use of a midlatitude summer atmosphere to represent the global average leads to an overprediction of between 4 and 8% in the radiative forcing. Differences between the UARS-based forcings and IPCC [1995] values are +17%, -6%, and -21% for CH₄, CCl₂F₂, and N₂O, respectively. The discrepancy for CCl₂F₂ is within the uncertainty (15%) associated with IPCC [1995] forcing estimates but slightly outside this bound for CH₄. It has been noted, however, that other studies have in fact indicated higher values (up to 20%) for the radiative forcing of CH₄ [IPCC, 1995], which is consistent with results presented here.

The reason for the large difference with IPCC [1995] in the computed N₂O forcing is unclear. The treatment of clouds may play a role; the clear-sky N₂O forcing at midlatitudes increases to 0.146 W m $^{-2}$ (see below). However, a systematic difference in cloud fractions or radiative properties would be expected to have the same sign for all three gases. Use of a midlatitude summer atmosphere with clouds only partially improves the agreement. The strong spectral overlap between CH₄ and N₂O may also be a factor in the discrepancy between N₂O forcings. Results presented here explicitly include the spectral overlap using a fixed abundance of 700 ppb CH₄. The N₂O forcing from IPCC [1995] is calculated based on an analytic fit to model calculations that includes an empirically determined correction for the spectral overlap.

Sensitivities of calculated forcing clouds and to vertical profiles of mixing ratio in the stratosphere are summarized in Table 2. As expected, forcings are larger for clear skies because absorbing gases at the tropopause and in the lower stratosphere see a warmer effective temperature below, leading to an enhanced greenhouse influence. Differences range between 28 and 36% at midlatitudes and are even more pronounced in tropics. The latter result arises due to the higher contrast in

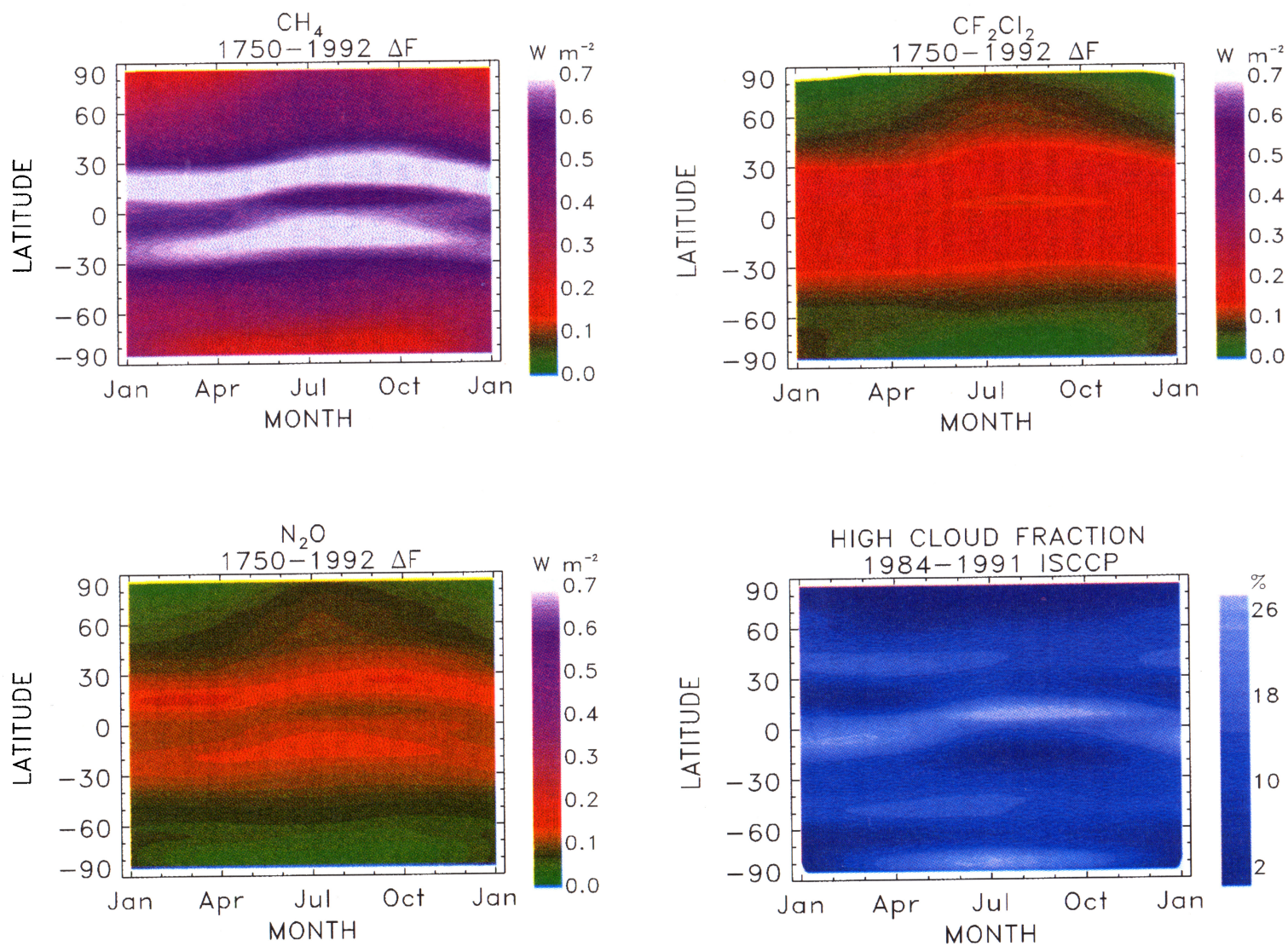


Plate 1. Seasonal and latitudinal dependence of the postindustrial radiative forcing calculated for CH_4 (upper left), (b) CCl_2F_2 (upper right), and N_2O (lower left). All forcings are in W m^{-2} . Also shown (lower right) is the distribution of high cloud fraction used in the calculations.

Table 1. Trace Gas Radiative Forcing

Species	Global Average	42°N Summer	IPCC [1995]
CH ₄	0.550	0.579	0.47
CCl ₂ F ₂	0.132	0.143	0.14
N ₂ O	0.111	0.115	0.14

Values are in $W m^{-2}$.

temperature between cloud top and the lower troposphere in the tropics as compared with midlatitudes. Cloud effects are similar to the 36% clear-sky enhancement for CCl₃F forcing at midlatitudes presented by Pinnock *et al.* [1995].

The impact of using realistic stratospheric profiles was discussed by Ramanathan *et al.* [1985] for CH₄ and N₂O and more recently by Hansen *et al.* [1997] and Christidis *et al.* [1997] for CFCs and HCFCs. However, previous studies either assumed exponential decreases using a constant scale height or used model-calculated profiles instead of observed distributions. Our calculations show a negligible impact on CH₄ and N₂O (Table 2), which is consistent with the results of Ramanathan *et al.* [1985]. The net IR flux at the tropopause does show a significant difference between the case of a uniform mixing ratio and one that decreases with height in the lower stratosphere. However, with the scaling procedures used here to approximate CH₄ and N₂O mixing ratios in the preindustrial stratosphere, this flux difference appears in both the preindustrial and contemporary cases, and very nearly cancels out in the radiative forcing.

On the other hand, the forcing for CCl₂F₂ is increased by up to 10% if a uniform mixing ratio is used to represent the contemporary atmosphere. One reason that this effect is larger than for CH₄ or N₂O is that the preindustrial atmosphere contained no CCl₂F₂; therefore the difference in net flux between the case of a uniform and realistic vertical profile of CCl₂F₂ translates directly to a difference in the computed radiative forcing. Furthermore, the opacity of the atmosphere is generally less for the CCl₂F₂ bands (9–11 μ m) in comparison with the major CH₄ (7.7 μ m) and N₂O (7.8 μ m) absorption bands. Consequently, changes in CCl₂F₂ abundances are felt over a greater range of altitudes, including the middle and upper stratosphere.

4.2. Lifetimes for CCl₂F₂ and N₂O

Instantaneous lifetimes were calculated using the sum of global loss rates for the four separate seasons (equation (4)) and the mean atmospheric burden during the same period (equation (3)). Lifetimes for CCl₂F₂ and N₂O are 114 ± 22 and 118 ± 25 years, respectively. Uncertainties are estimated based on the standard deviation of the CLAES zonal means where loss rates are a maximum (12% for CCl₂F₂ and 9% for N₂O), uncertainty in actinic flux (5% in solar irradiance and 10% in

atmospheric transmission), and absorption cross section uncertainties (10% for CCl₂F₂ and 15% for N₂O).

Calculated rates for stratospheric loss were largest in the tropical midstratosphere. Globally integrated rates were found to be moderately dependent on season; removal rates were 12% larger during the equinoxes as compared with the solstices. These higher rates are a consequence of smaller solar zenith angles in the tropics during the equinoxes, with associated increases in actinic fluxes in the lower stratosphere.

Previous estimates of the CCl₂F₂ lifetime range from 95 years [Ko *et al.*, 1991] to 180 years [Cunnold *et al.*, 1994]; the range for N₂O is 110 years [Ko *et al.*, 1991] to 182 years [Golombek and Prinn, 1986]. Our results favor the lower end of these ranges and are in good agreement with previous instantaneous lifetimes, 116 years for CCl₂F₂ and 123 years for N₂O, obtained by Minschwaner *et al.* [1993]. The latter estimates were obtained using the same empirical technique and nearly identical calculations for photochemical loss as used here; however, the level of agreement is somewhat surprising in view of the fact that the trace gas distributions used by Minschwaner *et al.* [1993] were based on a relatively sparse collection of balloon and aircraft data.

The steady state lifetime, where emissions to the atmosphere exactly balance photochemical loss, is generally shorter than the instantaneous lifetime for gases whose concentrations are increasing with time. The difference arises from the time lag between temporal changes in abundances for the stratosphere relative to the troposphere. The distinction between steady state and instantaneous lifetime is small for N₂O because the growth rate of about 0.25% year⁻¹ [Montzka *et al.*, 1992] does not significantly impact stratospheric/tropospheric abundances over timescales relevant for the turnover of stratospheric air (less than 5 years [Rosenlof and Holton, 1993]). However, the mean growth rate for CCl₂F₂ is much larger, averaging about 3.5% year⁻¹ between 1988 and 1992 [Elkins *et al.*, 1993]. Assuming a mean age of between 2 and 4 years for midstratosphere air in the tropics [Hall and Plumb, 1994; Boering *et al.*, 1996] and an instantaneous lifetime of 114 years, the estimated steady state lifetime for CCl₂F₂ is between 99 and 106 years. Including the uncertainty in instantaneous lifetimes yields steady state lifetimes of 103 ± 25 years for CCl₂F₂, and 117 ± 26 years for N₂O.

Table 2. Forcing Sensitivities, Summer Season

Species	Clear Sky		Fixed Strat ^a	
	42°N	3°N	42°N	3°N
CH ₄	29	36	1	1
CCl ₂ F ₂	36	51	8	10
N ₂ O	28	35	2	1

Values are percent increases above standard cases.

^aConstant trace gas mixing ratio above the tropopause.

Steady state lifetimes of 102 and 120 years for CCl_2F_2 and N_2O , respectively, are presented by IPCC [1995]. These lifetimes, based primarily on results of 2-D model calculations, agree very well with our UARS-based values. Results from the Goddard Institute for Space Studies/UCI three-dimensional chemical transport model yield steady-state lifetimes of 90 and 113 years for CCl_2F_2 and N_2O , respectively [Avallone and Prather, 1997], which are somewhat shorter than those calculated here but within the range of uncertainty. As noted by Avallone and Prather [1997], there may be a tendency in the model for excessive vertical mixing in the tropical stratosphere, leading to enhanced rates of calculated destruction. The use of observed tracer correlations by Volk *et al.* [1997] indicates lifetimes of 87 and 122 years for CCl_2F_2 and N_2O , respectively. These are also consistent with our results, although the CCl_2F_2 steady state lifetime is near the low range of the UARS-based value.

5. Conclusions

Our radiative calculations constrained by UARS and ISCCP observations imply a total postindustrial forcing of 0.79 W m^{-2} due to increasing burdens of trace gases CH_4 , CCl_2F_2 , and N_2O . For comparison, the estimated CO_2 -induced forcing is 1.56 W m^{-2} over the same period [IPCC, 1995]. Equally important, our results indicate a strong dependence of the trace gas forcing on season and latitude. At midlatitudes, forcings are largest during the summer-fall season, and seasonal amplitudes are about 10%. Latitudinal values of the trace gas forcing increase by nearly a factor of 3 between the winter high latitudes and subtropics.

Cloud effects, particularly the fraction of high clouds in the tropics, play a significant role in regulating the radiative forcing. Clouds reduce the clear-sky forcing by about 30% at midlatitudes and by up to 50% in the tropics. In this regard it is important to note that even small changes in cloud amount could be just as important as changes in trace gas abundances. In addition, the stratospheric distribution of CCl_2F_2 is important in determining the magnitude of its forcing. Since the stratospheric profile of CCl_2F_2 is determined in part by UV photolysis and opacity by O_3 , this implies a possibly important connection between the photochemical and climatic influence of this gas.

The radiative forcing and atmospheric lifetime both enter into the evaluation of the global warming potential (GWP) of a greenhouse gas. The GWP is defined as the time-integrated radiative forcing from the instantaneous release of 1 kg of trace gas, expressed relative to that from 1 kg of CO_2 [IPCC, 1990]. For the trace species considered here, the magnitudes of postindustrial radiative forcings and atmospheric lifetimes are broadly similar to currently accepted values, implying GWPs which are consistent with results presented by IPCC [1995]. However, our radiative forcing by N_2O is 21% smaller in magnitude, and our steady state lifetime is about 2% shorter; taken together, these results sug-

gests that the N_2O GWP may be about 23% less than the currently recommended value.

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