Consumption of atmospheric methane by soils: A process-based model

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Abstract. A process-based model for the consumption of atmospheric methane (CH_4) by soils was developed to identify the most important factors affecting uptake rates and to determine whether the current uncertainties in the estimated size of the global soil sink might be reduced. Descriptions of diffusion and microbial oxidation processes, which together determine the CH_4 flux, were included. The results suggest that the global sink strength lies within the range 20-51 Tg yr^{-1} CH_4, with a preferred value of 38 Tg yr^{-1} CH_4. Dry tropical ecosystems account for almost a third of this total. Here microbial activity rather than diffusion is limiting uptake. It is also in these areas that the impact of any intensification in agriculture will be the most pronounced, with a possible future reduction in uptake in excess of 3 Tg yr^{-1} CH_4. This is in contrast to the overall impact of global warming, which is expected to leave the size of the global soil sink relatively unchanged.

1. Introduction

Methane is currently present in the atmosphere at a concentration of about 1.72 ppmv [*Houghton et al.*, 1996], contributing an estimated 15% to anthropogenic global warming [*Rodhe*, 1990]. CH_4 is removed from the atmosphere largely through reaction with hydroxyl molecules (OH) in the troposphere [*Fung et al.*, 1991] and also as a result of microbial oxidation in soils. Although recent estimates of 15-45 Tg yr^{-1} CH_4 for the size of the global soil sink put it at only about 10% of that due to reaction with OH [*Houghton et al.*, 1995], the estimated excess of sources over sinks for CH_4, which accounts for the present rate of atmospheric increase, is of a similar magnitude. Therefore, changes in the strength of the soil sink could potentially have a significant effect on future atmospheric CH_4 concentrations and thus on the rate and extent of global warming.

Global estimates of trace gas fluxes are commonly made by extrapolation from field and laboratory studies; point measurements are scaled up, assuming them to be representative of all locations sharing some common classification such as soil or ecosystem type [*Matson et al.,* 1989; *Schimel and Potter*, 1995; *Steudler et al.*, 1989]. Global fluxes are then estimated by multiplying the representative flux by the area within each classification [*Dörr et al.*, 1993; *Ojima et al.*, 1993; *Potter et al.*, 1996]. However, the assumption that point measurements made at a limited number of locations and for a limited time period can really be representative of all locations over an extended time period can lead to significant errors [*Aselmann*, 1989; *Matson et al.*, 1989]. Such extrapolations are valuable as initial estimates and for defining maximum and minimum limits, but they cannot necessarily provide information about fluxes under novel conditions, particularly those arising as a result of climatic change.

An alternative approach is to construct a “process-based” model. Here the most important biogeochemical processes determining trace gas exchange are modeled explicitly, usually within a regular array of cells representing areas of the Earth’s surface. Soil, ecosystem, and meteorological data are provided by linking with suitable global databases. However, scale incompatibilities can still be a problem with this approach. Descriptions of trace gas processes are typically developed using single-location studies with characteristic scales of the order of a few centimeters. This is in contrast to their application over the entire globe, where characteristic scales can be over 100 km for a 1° by 1° grid near the equator. Process descriptions must therefore be reparameterized so as to be applicable at the coarser scale, which will inevitably result in a loss of accuracy and which may introduce significant errors if the processes involved respond in a nonlinear fashion [*Schimel and Potter*, 1995]. Use of a finer grid scale may reduce these errors, but the maximum resolution that can be employed is ultimately limited by the resolution of suitable data sets and also by consideration of the computational resources available. This type of model has been successfully applied to a number of trace gas processes, such as the production of N_2O [*Bouwman et al.*, 1993] and CO_2 [*Potter et al.*, 1993] by soils and CH_4 by wetlands [*Cao et al.*, 1996a] and rice paddies [*Cao et al.*, 1996b] and the consumption of CH_4 by soils [*Potter et al.*, 1996].

Consumption of atmospheric CH_4 by soils has been shown to be the result of an entirely biological oxidation process [*Bender and Conrad*, 1994; *Whalen and Reeburgh*, 1990]. The activity of methanotrophs (and thus the potential rate of CH_4 oxidation within the soil) is regulated by a number of environmental factors such as temperature, soil moisture, soil nitrogen content, organic matter content, and *p*H. However, the primary control on the flux of CH_4 into soils is usually considered to be the rate of diffusion of CH_4 within the topsoil [*Born et al.*, 1990; *Dörr et al.*, 1993], which is controlled by both the physical structure and moisture content of the soil. As a result, in fine-textured and/or relatively water-logged soils, low gaseous diffusivity tends to restrict uptake, and the flux of CH_4 into the soil is not significantly affected by the degree of microbial activity. Under conditions of high diffusivity, on the other hand, the rate of microbial oxidation is often limiting, especially in extremely dry soils when methanotrophs may experience a significant water stress. Between these two extremes, lies a regime where both effects will play a role in limiting CH_4 consumption [*Reeburgh et al.*, 1993]. Accurate determination of the flux in all possible soil types and environmental conditions will therefore require, in addition to an estimation of gaseous diffusivity, an explicit treatment of microbial activity. Despite this, previous regional and global estimates have tended to place most emphasis on the physics of soil diffusivity, while biological controls over CH_4 uptake have taken a minor role. The aim of this study, then, is to develop a mechanistic model for global consumption of CH_4 by soils, which incorporates descriptions of the most important controlling factors, related to both gaseous diffusion and microbial activity.

2. Model Implementation

2.1. Calculation of Soil CH_4 Flux

A comprehensive description of the uptake of atmospheric CH_4 by soils depends on the ability to model the depth profile of CH_4 oxidation rate within the soil from all the controlling factors. Unfortunately, not enough is known about the physiology of the methanotrophs responsible to be able to derive such a distribution. Additionally, the soil profile information available on a global basis is restricted to little more than grain size fractions, whereas knowledge of soil organic matter content, inorganic N, and *p*H would also be required. A simplification of the system is therefore essential.

As a result of soil diffusivity being frequently limiting to CH_4 uptake, *Potter et al.* [1996] devised a simple parameterization based on Fick's law of diffusion [*Monteith and Unsworth*, 1990]:

$$J_{\mathrm{CH4}} = D_{\mathrm{CH4}} \frac{\Delta C_{\mathrm{CH4}}}{\Delta z} \tag{1}$$

where J_{CH4} is the flux of CH_4 into the soil, D_{CH4} is the diffusivity of CH_4 in the topsoil ($\mathrm{cm}^2\ \mathrm{s}^{-1}$), and $\Delta C_{\mathrm{CH4}}\ /\ \Delta z$ is a fixed concentration gradient, taken to be 0.04 ppmv cm^{-1}, on the basis of regression analysis of several published soil CH_4 concentration profiles [*Born et al.*, 1990; *Whalen et al.*, 1991]. Although *Potter et al.* [1996] modeled gaseous diffusivity from the consideration of soil texture, temperature, and moisture content, no explicit account was taken of the influence that microbial responses to environmental forcings may have upon J_{CH4}.

An alternative scheme, which retains the ability to account for changes in microbial oxidation activity, is to assume a profile of the oxidation rate constant against depth taken from experimental determinations. However, calculation of the CH_4 flux is not trivial, and in general, there will be no analytical solution unless the profile can be described by a simple function of depth such as a constant [*Born et al.*, 1990; *Striegl*, 1993] or a negative exponential [*Dörr et al.*, 1993; *Striegl*, 1993]. Alternatively, a numerical solution for one-dimensional diffusion with first-order reaction kinetics based on the partitioning of the soil profile into discrete layers [*Kruse et al.*, 1996; *Moldrup et al.*, 1996a, b; *Striegl and Ishii*, 1989] is possible. Extensive testing with a series of compartmentalized models suggested that there was no significant advantage to be gained in terms of the accuracy of flux estimation by the use of a profile taken directly from observations over the use of a much simpler triangular- or pulse-shaped approximation. In undisturbed soils, oxidation activity tends to be concentrated at depths of 5 - 7 cm [*Adamsen and King*, 1993; *Bender and Conrad*, 1994; *Koschorreck and Conrad*, 1993; *Kruse et al.,* 1996; *Schnell and King*, 1994], although a few soils exhibit oxidation activity maxima significantly deeper than this [*Reeburgh et al.*, 1997]. Away from the maximum there is typically a rapid reduction in activity toward the surface and a slightly gentler decline with increasing depth. All oxidation activity was therefore assumed to take place at a single depth of 6 cm in the soil (i.e., as a delta-function representation). This parameterization has the advantage that the flux rate could be approximated analytically, with no need for a computationally intensive, multiple-layer scheme.

The flux of atmospheric CH_4 into the soil (J_{CH4}) is calculated on the basis that all CH_4 consumption takes place at a rate determined by a first-order oxidation rate constant k_d (s^{-1}) and at depth z_d (centimeters). In addition, all oxidized CH_4 is assumed to originate from the atmosphere, ignoring possible methanogenic production of CH_4 within the soil, either in anoxic microsites [*Conrad*, 1995; *Tate and Striegl*, 1993] or at great depth [*Striegl et al.*, 1992; *Whalen and Reeburgh*, 1990]. The model will therefore not be able to account for the effect on soil flux of a shift in the balance between methanogenic production and methanotrophic consumption within the soil in response to changing environmental conditions, which can be important [*Keller et al.*, 1993; *Yavitt et al.*, 1993]. In the steady state the diffusive flux into the soil described by Fick's law (equation (1)) must be equal to the rate of microbial oxidation given by

$$J_{\mathrm{CH4}} = k_d \times C_{d\mathrm{CH4}} \tag{2}$$

where $C_{d\mathrm{CH4}}$ is the concentration of CH_4 at depth z_d. If $C_{0\mathrm{CH4}}$ is defined as the atmospheric CH_4 concentration (ppmv), setting $\Delta z = z_d$ and $\Delta C_{\mathrm{CH4}} = C_{0\mathrm{CH4}} - C_{d\mathrm{CH4}}$ in (1) and equating (1) and (2) gives

$$D_{\mathrm{CH4}} \frac{(C_{0\mathrm{CH4}} - C_{d\mathrm{CH4}})}{z_d} = k_d C_{d\mathrm{CH4}} \tag{3}$$

Writing (3) in terms of $C_{d\mathrm{CH4}}$, substituting back into (1), and rearranging now gives

$$J_{\mathrm{CH4}} = \frac{C_{0\mathrm{CH4}} D_{\mathrm{CH4}}}{z_d} \left(1 - \frac{D_{\mathrm{CH4}}}{(D_{\mathrm{CH4}} + k_d z_d)} \right) \times F \tag{4}$$

where F is a factor (616.9 mg ppmv^{-1} cm^{-1} CH_4) chosen to give J_{CH4} in units of mg m^{-2} d^{-1} CH_4. Inspection of this formulation confirms that soil CH_4 uptake will not always be limited by gaseous diffusivity as is often assumed. Instead, to a first approximation, uptake depends on the ratio of diffusivity to methanotrophic activity, with uptake determined almost exclusively by diffusivity at high rates of microbial activity and by microbial activity at high diffusivities.

Although the atmospheric CH_4 concentration is observed to vary with both latitude and season [*Fung et al.*, 1991], for simplicity a fixed average value for $C_{0\mathrm{CH4}}$ of 1.72 ppmv [*Houghton et al.*, 1996] was adopted. This will result in errors of no more than 5%, even at high latitudes, which exhibit the greatest deviation of $C_{0\mathrm{CH4}}$ from the global mean.

Oxidation kinetics are assumed to remain first order over all possible soil CH_4 concentrations from typical atmospheric values down to zero. In practice, although oxidation is observed to continue at mixing ratios of much less than 0.1 ppmv [*Bender and Conrad*, 1992, 1993; *Koschorreck and Conrad*, 1993; *Whalen et al.*, 1990], kinetics diverge from true first order below a threshold of about 0.3 ppmv [*Bender and Conrad*, 1993]. Considering that soils in which $C_{d\mathrm{CH4}}$ is likely to fall to below this threshold will be characterized by very low diffusivities and thus tend to have a relatively small flux, on average, errors due to the assumption of pure first-order kinetics will not be significant.

2.2. Calculation of Soil CH_4 Diffusivity

The gaseous diffusivity of CH_4 within the soil (D_{CH4}) is described by

$$D_{\mathrm{CH4}} = G_{\mathrm{soil}} \times G_T \times D_{0\mathrm{CH4}} \tag{5}$$

where $D_{0\mathrm{CH4}}$ is the diffusivity of CH_4 in free air, taking a value at STP of 0.196 cm^2 s^{-1} [*Cussler*, 1984], and G_{soil} and G_T are scalars accounting for the effects of soil structure and temperature respectively. It has been suggested that the aqueous diffusion of CH_4 in soil particle water films may play an important limiting role in uptake [*Castro et al.*, 1995; *Koschorreck and Conrad*, 1993]. For simplicity, liquid phase transport has not been explicitly modeled in this study, although it is likely that the qualitative influence of soil moisture on uptake rate due to aqueous diffusion will be accounted for through changes in modeled gaseous diffusion. A distinct advantage of the diffusion-consumption flux calculation scheme (equation (4)) over the fixed gradient approach (equation (1)) is that any error in the estimation of diffusivity will be partly compensated for: an overestimation of D_{CH4} will lead to a reduced concentration gradient in the soil, thereby reducing the resulting overestimation of J_{CH4}, and vice versa.

2.2.1. Soil structural dependence. A number of different schemes for calculating gaseous diffusivity in porous media, such as soils, have been proposed. Some require the fitting of parameters from experimental studies on individual soils [*Nielson et al.*, 1984; *Troech et al.*, 1982] and so are unsuitable for global implementation. Others developed for more general application are derived by a "cut-and-random-rejoin" treatment and use estimates of total and air-filled porosity together with a tortuosity factor [*Millington and Quirk*, 1961; *Millington and Shearer*, 1971]. We adopt here the new parameterization of *Moldrup et al.* [1996b], with the scalar G_{soil} defined as

$$G_{\mathrm{soil}} = \Phi^{4/3}\left(\frac{\varepsilon}{\Phi}\right)^{1.5 + 3/b} \tag{6}$$

where Φ is the total pore volume (cm^3 cm^{-3}), ε is the air-filled porosity (cm^3 cm^{-3}), and b is a constant that can be empirically predicted from soil texture [*Saxton et al.,* 1986]. This parameterization gives a good prediction of the increase in diffusivity with soil-air content, and it has been found to be superior to the commonly used scheme of *Millington and Quirk* [1961] in a number of soils [*Moldrup et al.*, 1996b; *Olesen et al.*, 1996]. It is also mathematically simple, does not rely on the use of pore size classes, and has parameters that may be estimated from widely available soil characteristics.

2.2.2. Temperature dependence. The diffusivity of CH_4 in free air is a function of temperature, with G_T defined after the linear approximation of *Potter et al.* [1996]:

$$G_T = 1.0 + 0.0055T \tag{7}$$

where T is the temperature in degrees Celsius.

2.2.3. Surface air turbulence effects. The effect of wind action in increasing the rate of mass transfer of CH_4 is unlikely to extend any deeper than 1 or 2 cm, even in very coarse soils [*Farrell et al.*, 1966; *Scotter et al.*, 1967]. For most other soils the diffusion distance of 6 cm is likely to be an order of magnitude greater than the depth of influence of wind turbulence. This effect will therefore play only a very minor role in determining soil flux, and so it has been omitted.

2.3. Calculation of CH_4 Oxidation Activity

The CH_4 oxidation rate constant k_d is defined by

$$k_d = r_N \times r_{sm} \times r_T \times k_0 \tag{8}$$

where k_0 is the base oxidation rate constant for an uncultivated moist soil at 0°C and r_N, r_{sm}, and r_T are adjustment factors accounting for the effects of agriculture, suboptimal soil moisture conditions, and temperature, respectively. The *p*H has been shown to be an important factor at elevated CH_4 levels [*Dunfield et al.*, 1993], with a clear optimum for methanotrophic activity. However, studies performed at ambient pCH_4 are much less conclusive [*Hütsch et al.*, 1994; *Yavitt et al.*, 1993], while *Dörr et al.* [1993] could find no significant correlation between *p*H and CH_4 uptake rate in a wide range of soils. Furthermore, as there is currently no global data set for topsoil *p*H, it was necessary to omit the influence of *p*H from the model description.

Reported peak oxidation rate constants in nonagricultural soils range from under 2×10^{-4} s^{-1} to over 5.0×10^{-3} s^{-1} [*Adamsen and King*, 1993; *Bender and Conrad*, 1994; *Koschorreck and Conrad*, 1993; *Schnell and King*, 1994], with equivalent agricultural soils typically exhibiting about 30% of this [*Bender and Conrad*, 1994;

Koschorreck and Conrad, 1993]. A value for k_0 was obtained by fitting calculated fluxes generated using equation (4) together with measured values of gaseous diffusivity to observed values, correcting for soil temperature (where reported), as is shown in Table 1. A k_0 value of 8.7×10^{-4} s^{-1} ($R^2 = 0.55$) was obtained, which is consistent with the reported range of rate constants. Use of a fixed gradient scheme resulted in a considerably poorer fit to the observed J_{CH4} data, with an optimized gradient of 0.039 ppmv cm^{-1} giving an R^2 of only 0.08.

2.3.1. Temperature response. A number of studies have reported temperature responses of atmospheric CH_4 consumption both in situ [*Born et al.*, 1990; *Castro et al.*, 1995; *Crill*, 1991; *Koschorreck and Conrad*, 1993] and with soil samples incubated in the laboratory [*Crill et al.*, 1994; *King and Adamsen*, 1992; *Whalen and Reeburgh*, 1996]. The measured temperature dependence of atmospheric CH_4 uptake is profoundly influenced by the diffusive regime, particularly for measurements made in situ and on undisturbed cores, where there can be virtually no temperature response at low diffusivities and a pronounced response at high diffusivities [*Mosier et al.*, 1996]. Any determination of temperature dependence where diffusion is limiting to some degree will therefore not elucidate the underlying effect upon methanotrophic behavior.

Increasing microbial activity with temperature can typically be described by a function of the form $Q_{10}^{(T/10)}$, where Q_{10} is the 10° temperature quotient. Incubation studies at elevated CH_4 mixing ratios with mainly landfill cover soils have found Q_{10} values over temperatures in the range 0° - 20°C of 1.8 - 2.0 [*Boeckx et al.,* 1996], 1.4 - 2.1 [*Dunfield et al.*, 1993], and 1.8 - 2.1 [*Whalen et al.*, 1990]. Although *Crill et al.* [1994] found a Q_{10} value of only 1.3 in incubation experiments at 10 ppmv, their samples were considerably larger and more moist, so it seems quite likely that diffusion was still limiting. A Q_{10} value of around 2.0 is consistent with typical values for enzymatic activity [*Atlas and Bartha*, 1987] and also with values adopted in other trace gas flux modeling studies [*Cao et al.*, 1996a; *Potter et al.*, 1993; *Parton et al.*, 1996; *Schimel and Potter*, 1995]. Despite there being no reliable Q_{10} results for methanotrophic activity under atmospheric CH_4 concentrations, considering that a Q_{10} value of 2.0 appears to have such general microbiological applicability [*Schimel and Potter*, 1995], it was adopted here. After a maximum has been reached in the region of 25° - 31°C, activity rapidly declines [*Boeckx and van Cleemput*, 1996; *Boeckx et al.*, 1996; *Whalen et al.*, 1990; *Whalen and Reeburgh*, 1996]. The complete temperature response is thus of the form shown in Figure 1.

Some studies report significant CH_4 oxidation occurring at temperatures down to a couple of degrees below zero [*Castro et al.,* 1995; *Crill et al.*, 1994; *King and Adamsen*, 1992], unless the soil is frozen, when CH_4 uptake virtually ceases [*Castro et al.*, 1995; *Crill*, 1991; *King and Adamsen*, 1992; *Koschorreck and Conrad*, 1993]. In this study, when monthly mean temperatures fall below 0°C, the surface layers of the soil are assumed to freeze, so that r_T is then set to zero.

The scalar r_T is therefore defined as

$$r_T = 0 \qquad T < 0°\mathrm{C} \qquad (9a)$$

$$r_T = \exp[0.0693T - 8.56 \times 10^{-7} T^4] \qquad T \geq 0°\mathrm{C} \qquad (9b)$$

where T is the temperature in degrees Celcius. The function for $T \geq 0°\mathrm{C}$ was chosen to have a Q_{10} response of 2.0 over the interval

0°-15°C and to produce an approximate fit to experimental measurements [*Dunfield et al.*, 1993; *Whalen et al.*, 1990; *Whalen and Reeburgh*, 1996].

2.3.2. Cultivation response. Nitrogen input to the soil as a result of agricultural fertilizer application, particularly when in an inorganic form, has been found to have a profound inhibitory effect on CH_4 oxidation activity [*Castro et al.*, 1995; *Hütsch et al.*, 1993; *King and Schnell*, 1994; *Schnell and King*, 1994; *Steudler et al.*, 1989], although the mechanism by which this inhibition operates has still not been fully determined. Typically, it can reduce soil uptake by the order of 60%, but there is wide variability, with a complete cessation of uptake being reported in some cases [*Hütsch et al.,* 1993]. The underlying microbial response to N application is difficult to deduce from the various studies to date, primarily because of the likely differences in the degrees of diffusive control on uptake between studies. Reduction in methanotroph activity as a result of land conversion for agriculture, due to both the application of N-based fertilizers and reduced soil organic matter content [*Hütsch et al.*, 1994], is therefore parameterized rather simply, according to the intensity of cultivation. Oxidation activity in land under 100% cultivation is assigned a multiplication factor of 0.25, chosen to enable the model to reproduce the range of reported cultivation-related reductions in consumption when typical soil diffusivities are taken into account. The scalar r_N is therefore defined as

$$r_N = 1.0 - (0.75 \times I_{\text{cult}}) \tag{10}$$

where I_{cult} is the fractional intensity of cultivation [*Matthews*, 1983]. Reduction in CH_4 uptake due to physical changes in soil structure arising out of land conversion [*Dobbie et al.*, 1996; *Hütsch*, 1996; *Keller et al.*, 1993] is implicit within this factor. No attempt has been made to account for the effect of anthropogenically enhanced atmospheric nitrogen deposition, which has been estimated to reduce CH_4 fluxes by about 30% in urban areas [*Goldman et al.*, 1995], as its global distribution is not well known.

2.3.3. Soil moisture response. Low moisture content has been observed to suppress CH_4 uptake by soils [*Mosier et al.*, 1996; *Nesbit and Breitenbeck*, 1992; *Torn and Harte*, 1996; *Whalen et al.*, 1990]. However, the lack of sufficient soil textural and hydrological information in many of these studies makes it difficult to compare the various quoted thresholds of soil moisture content in terms of water potential. In addition, as the soil water balance submodel used here makes no attempt to predict soil drying beyond wilting point, when inhibition would be greatest, a general scheme for soil microbial activity was adopted. The function describing the soil moisture scalar r_{sm} was adapted from *Potter et al.* [1993]:

$$r_{sm} = 1.0 \qquad (P + SM)/ETp > 1.0 \tag{11a}$$

$$r_{sm} = (P + SM)/ETp \qquad (P + SM)/ETp \le 1.0 \tag{11b}$$

where P is the monthly precipitation, SM is the soil moisture stored to a depth of 30 cm (or to the rooting depth if less), and ETp is the potential evapotranspiration. Some studies also limit soil microbial activity at high soil moisture contents [*Parton et al.*, 1996; *Potter et al.*, 1993], when oxygen supply would be restricted. However, the microaerophilic nature of methanotrophic bacteria [*Hanson*, 1980] makes this addition unnecessary.

2.4. Global Application

To estimate the size of the global soil sink for CH_4, the diffusion-consumption submodel was applied to a 1° by 1° grid. A mask was constructed for areas where consumption was certain to be negligible, initially defined by areas of water (including ocean and lakes) and land ice [*Matthews*, 1983]. Areas defined as desert were also masked out [*Matthews*, 1983], assuming the consumption of atmospheric CH_4 to be associated with the production of CO_2 within the soil [*Dörr et al*., 1993] and thus with the presence of vegetation. Although wetland soils can become net consumers of atmospheric CH_4 when the water table falls in dry seasons [*Harriss and Sebacher*, 1982] or by land drainage [*Martikainen et al.*, 1995], consumption of atmospheric CH_4 is generally a property of aerobic soils. For this reason, areas of permanent wetland were masked out according to their distribution and coverage [*Matthews and Fung*, 1987].

Average monthly precipitation and near-surface air temperature were taken from *Legates and Willmott* [1992]. Soil temperature at a depth of 6 cm was assumed to follow air temperature. This is a reasonable assumption given that the damping depth for annual air temperature cycles, even in poorly conducting peaty soils, is greater than 1 m [*Monteith and Unsworth*, 1990] and gives a minimum 95% of the surface amplitude at the depth where oxidation activity is concentrated.

The structure of the complete global model is shown schematically in Figure 2.

2.4.1. Derivation of soil properties. Soil physical properties required by the water balance and gaseous diffusivity calculations were estimated from soil particle size fractions after *Saxton et al.* [1986]. Soil particle size information consisted of two sets of data, describing percent sand and clay content, both derived from a global soil properties database [*Webb et al.*, 1992]. Since the average soil properties over the depth affecting diffusion will not necessarily be representative of those over the entire rooting depth, one set of data was averaged over the diffusion depth (6 cm) for use in gaseous diffusivity calculations, while another was averaged over 150 cm for use in the soil water balance submodel.

2.4.2. Soil water balance. The soil water balance submodel was based on the widely used "bucket" formulation, where changes in stored moisture arise out of imbalances between gains (precipitation) and losses (evapotranspiration and surface runoff) to the system [*Bouwman et al.*, 1993; *Potter et al.*, 1993; *Vörösmarty et al.*, 1989]. The scheme used here is similar to that of *Potter et al.* [1993] and so will not be discussed in detail. The main differences were the use of a simpler soil drying curve (representing the resistance of an already drying soil to further moisture loss) and the estimation of rooting depth (for use in calculating soil water storage capacity), which was initially assigned following a scheme based on vegetation type [*Webb et al.*, 1991] and modified according to cultivation intensity [*Matthews*, 1983] and soil profile thickness [*Webb et al.*, 1992]. Any precipitation added to the system in excess of the soil's field capacity was assumed to be lost as runoff with no transfer of moisture between grid cells. This is likely to result in an overestimation of CH_4 uptake during periods of intense rainfall, when soil water content may exceed field capacity, restricting gaseous diffusivity to near zero.

The water balance submodel was initialized by setting the soil to a uniform moisture ratio of 50% for all months. It was then run repeatedly until the average yearly soil moisture converged to within 0.01% between successive years.

3. Results and Discussion

3.1. Model Validation

3.1.1. Diffusion-consumption submodel. The full range of predicted gaseous diffusivity (0.001-0.04 $cm^2\ s^{-1}$) and oxidation rate constant (< 0.0001-0.0036 s^{-1}) allows for a range in CH_4 uptake of <0.01-2.45 mg $m^{-2}\ d^{-1}$ CH_4. This covers the bulk of typical measurements [*Dörr et al.,* 1993], although a number of studies have reported fluxes in excess of 2.5 mg $m^{-2}\ d^{-1}$ CH_4 [*Adamsen and King*, 1993; *Crill*, 1991; *Steudler et al.*, 1989], which cannot be explained by the current model configuration.

The diffusion-consumption scheme (equation (4)) was validated against two of the weekly average CH_4 uptake time series presented by *Mosier et al.* [1996], from which soil temperature, water content, and flux data were sampled (N = 30). For the first of the Mosier et al. time series, our scheme gave an R^2 value of 0.35, whereas none of the observed variance could be explained using a fixed gradient approach. However, our scheme performed very poorly against the second time series, although once measurements for which there was evidence of a significant inhibition of methanotrophic activity through water stress were removed, and allowance was made for the fact that CH_4 oxidation at this site appeared to be N-inhibited, an R^2 value of 0.05 was obtained. This was still an improvement over the fixed gradient method.

3.1.2. Global model. Global models where the characteristic spatial and temporal scales are substantially different from those of experimental measurements are virtually impossible to validate satisfactorily. However, as discussed in section 3.2, the model predictions are in broad agreement with global and regional estimates made by other methods.

3.2. Basic (Biological) Model

The size of the global soil sink of atmospheric CH_4 (J_{tot}) is estimated at 37.8 Tg yr^{-1} CH_4 by our model. This figure lies comfortably within the Intergovernmental Panel on Climate Change (IPCC) estimate of 15-45 Tg yr^{-1} CH_4 [*Houghton et al.*, 1996]. It is also consistent with the range of possible values of 5.6-58.2 Tg yr^{-1} CH_4 proposed by *Born et al.* [1990], and with the best estimate of *Dörr et al.* [1993] of 28.7 Tg yr^{-1} CH_4 (range 9.0-55.9 Tg yr^{-1} CH_4) made on the basis of a soil texture parameterization derived from an extensive survey of mainly Central European soils. The estimate presented here does, however, differ significantly from the model-derived estimate of *Potter et al.* [1996] of 17 Tg yr^{-1} CH_4. The primary reason for this is their assumption of a fixed soil CH_4 concentration gradient set at a relatively low value of 0.04 ppmv cm^{-1}. As they point out, this is a very conservative estimate which cannot explain fluxes larger than 1.0 mg $m^{-2}\ d^{-1}$ CH_4, which are widely observed in soils. Using a higher gradient of 0.07 ppmv cm^{-1}, they reported a figure of nearly 30 Tg yr^{-1} CH_4, which is closer to the findings of this present study.

The global distribution of CH_4 uptake is shown in Plate 1a. Uptake averaged by aggregated Holdridge life zone [*Leemans*, 1992] is detailed in Table 2. Dry tropical ecosystems tend to dominate as a result of the high diffusivity of the dry sandy soils characteristic of these areas, in association with high temperature driven microbial activity. However, this is in contrast to experimental studies that suggest that annual fluxes in tropical areas are generally much lower [*Keller et al.*, 1993]. This is because only the oxidation of CH_4 in soils is considered, which will predict a low rate of consumption during periods of soil waterlogging owing to restricted gaseous diffusivity. Flux measurements, on the other hand, can register a net emission of CH_4 if the soil becomes anaerobic. Annual uptake based on flux measurements may then be quite small or even negative, with net emission during wet seasons offsetting consumption at other times of the year.

3.2.1. Effects of microbial water stress. The effect of the scheme for the estimation of microbial water stress adopted here was to suppress J_{tot} by over 16% from a maximum of 45.2 Tg yr^{-1} CH_4. The model is therefore relatively sensitive to this choice, and the use of a considerably more complex surface scheme that is able to predict the drying of surface layers beyond the permanent wilting point would probably be justified.

3.2.2. Effects of agriculture. The effect of land conversion for agriculture and particularly the use of inorganic nitrogen fertilizers was to reduce J_{tot} by 3.7 Tg yr^{-1} CH_4 from a potential 41.5 Tg yr^{-1} CH_4. *Potter et al.* [1996] estimated the global effect of agriculture at around 13%, which is not dissimilar from the 9% reduction predicted here. The effect on J_{tot} of the degree to which agricultural land use reduces CH_4 oxidation activity in the soil is shown in Figure 3, and the effects of cultivation by ecosystem classification are shown in Table 2. *Dobbie et al.* [1996] estimated a reduction of 2.3 Tg yr^{-1} CH_4 due to land conversion to agriculture in the temperate zone, while *Ojima et al.* [1993] estimated that it could be as high as 6 Tg yr^{-1} CH_4. This present study puts the figure rather lower at 1.7 Tg yr^{-1} CH_4, some 19% of the potential (undisturbed) soil sink in the temperate region.

Future intensification of agriculture, particularly in currently nonintensively cultivated dry tropical regions, can be expected to have a significant impact. The effect of the suppression of methanotrophic activity in temperate areas by cultivation is restricted by the predominantly diffusion-limited nature of the processes in these soils. In much warmer and drier regions where diffusion may not generally be limiting, the full impact on uptake of a reduction in methanotrophic activity can potentially be realized. An equivalent intensity of cultivation would therefore be likely to produce far more than the average 19% temperate zone reduction. If all areas defined as having a cultivation intensity ≥20% [*Matthews*, 1983] are set to 100% (representing global application of intensive agriculture), then J_{tot} falls by almost 5 Tg yr^{-1} CH_4 to 33.2 Tg yr^{-1} CH_4, of which over 3 Tg yr^{-1} CH_4 occurs in dry tropical regions.

3.2.3. Seasonal variations in CH_4 consumption. Figure 4 shows the seasonality in CH_4 consumption averaged by latitude zone. As would be expected from consideration of climatic factors alone, zones nearest the equator (±30°) show very little seasonality in CH_4 uptake, whereas higher latitudes have a much more pronounced response, with an almost total cessation of consumption during cold winter months in the northern hemisphere. Contrasting this with the seasonality in consumption calculated with the temperature dependence of methanotrophic activity removed (not shown), suggests that much of the seasonal control on CH_4 uptake at midlatitudes and high latitudes is due to changes in diffusivity within the soil arising from changing soil moisture content. However, temperature-driven variations in microbial activity appear to restrict the potential summer maximum at high northern latitudes (90° - 60°N) by over 25%, and they significantly enhance the corresponding trend in the response of southern midlatitudes (30° - 60°S).

3.3. Fixed Soil CH_4 Gradient Model

As a comparison of flux estimation schemes, our model was adjusted to use a fixed CH_4 gradient in the soil. A value of 0.087 ppmv cm^{-1} was chosen to generate a similar value for J_{tot} (38.0 Tg yr^{-1} CH_4) as the basic model. The result of this was a global distribution as shown in Plate 1b. The very pronounced contrast in uptake at similar latitudes is due primarily to variations in soil texture, and in general, low CH_4 uptake correlates strongly with regions of high-percent clay soils (not shown).

The differences in flux distribution between the biological and fixed gradient models (i.e., Plates 1a and 1b, respectively) arise out of the sole control over CH_4 uptake of gaseous diffusivity in the fixed gradient model. This highlights the regions where environmental conditions are such that microbial oxidation activity is strongly limiting to uptake. The impact of agricultural land use (mainly through the inhibition of methanotrophic activity by N-based fertilizers) can be seen, particularly in central North America. The effects of microbial water stress in restricting uptake in regions bordering areas of permanent desert are also apparent. It should be noted that these are areas where soils are relatively coarse, and thus soil diffusivity is not limiting. Conversely, there is very little effect on uptake of the intensive cultivation in much of Europe, where soils tend to be relatively fine, and thus uptake is largely limited by the resulting low soil diffusivity. Also apparent in the spatial contrast between the two models is the substantial underestimation by the fixed gradient model of uptake in low-diffusivity clay soils (e.g., South America, equatorial Africa, and Southeast Asia). As a result, the importance of soil texture in determining the spatial flux distribution appears to be much less in the biological model.

3.4. Sensitivity Analysis

3.4.1. Model parameters. Probably the most critical single parameter in the model, and the one for which there is the least confidence, is the base oxidation rate constant k_0. The sensitivity of global uptake to the value of this constant is shown in Figure 5. It is apparent that although J_{tot} is relatively sensitive to k_0 at low values, in general, the choice of k_0 will be significantly less critical than would be the choice of $\Delta C / \Delta z$ in a fixed gradient calculation scheme, where J_{tot} is directly proportional to $\Delta C / \Delta z$. Assuming the confidence limits in k_0 to be the maximum and minimum values which still achieve a fit to the flux data of Table 1 with $R^2 > 0$, a likely range for J_{tot} is 20 - 51 Tg yr^{-1} CH_4.

3.4.2. Choice of gaseous diffusivity scheme. Use of the *Millington and Shearer* [1971] gaseous diffusivity scheme resulted in a slightly higher estimate for J_{tot} of 42.4 Tg yr^{-1} CH_4. However, the *Millington* [1959] and *Millington and Quirk* [1961] schemes both resulted in much lower estimates of 24.4 and 25.0 Tg yr^{-1} CH_4, respectively, which is not unexpected given their tendency to underpredict gaseous diffusivity [*Davidson and Trumbore*, 1995; *Moldrup et al.*, 1996b]. A degree of care must therefore be taken in the choice of scheme. One deficiency in the coupling of the gaseous diffusivity and soil water balance schemes is that soil diffusivities much greater than 0.04 cm^2 s^{-1} are not predicted, although they have been observed in the field [*Dörr et al.*, 1993]. The reason for this is likely to be the artificial restriction of the minimum soil water potential to wilting point. It is also possible that the presence of macropores, such as worm and root channels and cracks from wetting-drying cycles, are important in generating the highest observed diffusivities, although the incorporation of these effects would then require consideration of the macropore structure of the soil, which would be extremely complex.

3.4.3. Climatic change. The effect of changes in global temperatures on J_{tot} was initially investigated in a rather crude manner by uniformly incrementing all of the monthly average temperature values by between 0.5° and 5.0°C. Soil moisture was not allowed to respond to this change. The response of J_{tot} is shown in Figure 6. Initially, increases at high latitudes dominate the global response (not shown), but beyond about 1° of warming, declining uptake rates at low latitudes start to determine the overall response, which is a result of the steep decline in microbial activity beyond the peak at 25° - 30°C (Figure 1). Although the response of J_{tot} could be expected to be different with a more realistic global distribution of temperature increase and/or an alternative microbial temperature response function, it seems likely that J_{tot} is relatively insensitive to the direct effects of temperature change during global warming. If average monthly soil moisture content is now allowed to decrease in response to increasing air temperature (still with no precipitation changes accounted for), then J_{tot} is unlikely to exceed about 41.5 Tg yr^{-1} CH_4 (Figure 6). Obviously, the highly simplified nature of the water balance scheme together with the crude climate change forcing applied mean that little weight can be attached to such estimates. Despite this, an increase in global CH_4 sink strength with global warming of about 1 Tg yr^{-1} $°C^{-1}$ CH_4 can probably be taken as a reliable upper limit.

An alternative method for estimating the possible impacts of climate change is to hybridize process-based and flux extrapolation approaches. In this case, average modeled fluxes for each Holdridge life zone [*Leemans*, 1992] are kept constant, while the change in the area of each zone under a global warming is considered. Average zonal fluxes were therefore applied to the estimated area of each Holdridge life zone under a ×2 CO_2 scenario [*Leemans*, 1990], giving a slight increase in J_{tot} of 1.7 to 39.5 Tg yr^{-1} CH_4.

4. Conclusions

The estimate of 37.8 Tg yr^{-1} CH_4 presented here for the size of the global atmospheric methane soil sink is consistent with most previous estimates. This study supports the current IPCC range of 15 - 45 Tg yr^{-1} CH_4 [*Houghton et al.*, 1996] but offers no justification for any downward revision of this to 15 - 30 Tg yr^{-1} CH_4, as proposed by *Potter et al.* [1996]. The size of the global soil sink appears surprisingly insensitive to the effects of global warming. It is likely that the impact of a ×2 CO_2 scenario will result in a change in sink strength within the range -1 to +3 Tg yr^{-1} CH_4. This is a smaller change than that which might arise as a consequence of any significant future intensification of agriculture.

Considering the importance of dry tropical ecosystems, which account for over 28% of the global sink from only 18% of the land area, and the paucity of experimental measurements there, considerably more research in these regions is highly desirable. In contrast, temperate ecosystems, which from a similar land area account for only 19% of the soil sink, have tended to receive most of the research effort.

To further improve process-based model descriptions of the consumption of CH_4 by aerobic soils, future experimental studies on microbially mediated factors affecting uptake rates are required, with reference to the degree to which gaseous diffusivity is limiting to that uptake. Ideally, in such studies, determinations of the gaseous diffusivity in the topsoil should be included. Allowance can then be made for the control on uptake exerted by the diffusion rate, and the underlying methanotrophic behavior can be determined. Diffusivity measurements in the field, though, are both difficult and cumbersome [*Davidson and Trumbore*, 1995], so that it is not surprising that they are rarely made alongside flux measurements. An acceptable compromise would be for the routine analysis of soil properties and hydrological state to be performed, which would allow estimates of soil diffusivity to be made via standard calculation schemes.

The next major step in CH_4 flux modeling should be to take a less reductionist approach. This current study has focused purely on the consumption of atmospheric CH_4, with no account taken of CH_4 production within the soil. In a similar manner, previous emission models have taken no account of the possibility of net consumption of atmospheric CH_4. An integrated CH_4 consumption and production model is therefore required in order to address the contribution of soils as a whole (both aerobic and anaerobic) to the atmospheric CH_4 budget. In fact, there is no reason why a sufficiently comprehensive soil scheme, able to account for both aerobic and anaerobic conditions and processes, could not be utilized to estimate the atmospheric exchange of CO_2 and N_2O in addition to CH_4, all as part of a single global model. However, for this to be achievable, substantial progress is required in the construction of data sets of global soil properties (both physical and chemical) at an increased vertical and horizontal resolution.

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Figure 1. Scalar function r_T for the effect of temperature on methanotrophic CH_4 oxidation activity (equation (9b)). Also shown are experimental data for CH_4 oxidation rate, scaled to a value of unity at $T = 0°C$, from *Dunfield et al.* [1993], *Whalen et al.* [1990], and *Whalen and Reeburgh* [1996].

Figure 2. Schematic representation of the global atmospheric methane soil consumption model. Shaded and unshaded boxes represent input data sets and calculation processes, respectively. The soil water balance and diffusion-consumption submodels are delineated by dashed lines.

Figure 3. Sensitivity of the annual global uptake of atmospheric CH_4 by soils (J_{tot}) to the degree of inhibition of methanotrophic oxidation activity resulting from the conversion of land for agriculture.

Figure 4. Seasonality of the monthly uptake of atmospheric CH_4 by soils, averaged by 30° latitude zones.

Figure 5. Sensitivity of the annual global uptake of atmospheric CH_4 by soils (J_{tot}) to the choice of value for base oxidation rate constant k_0.

Figure 6. Sensitivity of the annual global uptake of atmospheric CH_4 by soils (J_{tot}) to a uniform increase in average monthly temperatures across the globe, with fixed (solid circles) and variable (solid squares) monthly soil moisture.

Plate 1. Global distribution of annual uptake of atmospheric CH_4 by soils predicted by (a) the basic model and (b) the fixed 0.087 ppmv cm^{-1} soil gradient model.

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Table 1. Measured Soil Methane Fluxes J_{CH4} Compared With Values Calculated With the Diffusion-Consumption Scheme Using Experimentally Determined Values of CH_4 Diffusivity D_{CH4} and Temperature T and an Optimized Value for the Base Oxidation Rate Constant k_0

D_{CH4}, cm^2 s^{-1}	T, °C	J_{CH4} Observed, mg m^{-2} d^{-1} CH_4	J_{CH4} Calculated, mg m^{-2} d^{-1} CH_4	Reference
0.064	12.5[a]	2.9	1.79	*Dörr et al.* [1993]
0.032	12.5[a]	1.5	1.54	*Dörr et al.* [1993]
0.058	12.5[a]	2.9	1.76	*Dörr et al.* [1993]
0.0105	12.5[a]	0.35	1.02	*Dörr et al.* [1993]
0.0025	12.5[a]	0.23	0.36	*Dörr et al.* [1993]
0.036	0.0[a]	0.7	0.80	*Dörr et al.* [1993]
0.069[b]	10.0[a]	1.7	1.57	*Striegl* [1993]
0.043	14.0	0.9[c]	1.78	*Koschorreck and Conrad* [1993]
0.029	23.0	2.0	2.08	*Koschorreck and Conrad* [1993]
0.041[b]	4.5	0.55	1.06	*Whalen et al.* [1992]
0.036[b]	4.1	0.22	1.01	*Whalen et al.* [1992]
0.095[b]	1.5	0.62	0.96	*Whalen et al.* [1992]
0.048[b]	2.7	0.55	0.97	*Whalen et al.* [1992]

Diffusion-consumption scheme is given by equation (4). Value for the base oxidation rate constant k_0 was 8.7×10^{-4} s^{-1}.

[a] Estimated temperature.

[b] Estimated from topsoil air-filled porosity.

[c] Average flux.

Table 2. Methane Consumption by Soils, Averaged by Aggregated Holdridge Life Zone, for Both the Reference Case and a No Agriculture Scenario, Together With Percentage Reduction in Uptake Due to Cultivation

Aggregated Holdridge Life Zone		Flux Rate, mg m^{-2} yr^{-1} CH_4			Annual Sink, Tg yr^{-1} CH_4		
Class	Description	Reference	No Agriculture	Area, 10^6 km^2	Reference	No Agriculture	Cultivation-Related Reduction in Uptake, %
1	Tundra	100.2	100.3	10.5	1.05	1.05	0.0
2	Cold Parklands	207.0	210.3	2.8	0.59	0.59	0.0
3	Forest Tundra	161.0	161.8	8.9	1.44	1.45	0.7
4	Boreal Forest	187.3	196.4	15.2	2.85	2.99	4.7
5	Cool Desert	265.0	274.2	4.0	1.07	1.11	3.6
6	Steppe	255.9	316.9	7.4	1.91	2.36	19.1
7	Temperate Forest	240.6	297.3	10.0	2.41	2.98	19.1
8	Hot Desert	179.2	183.4	20.9	3.74	3.83	2.3
9	Chapparal	343.9	429.9	5.6	1.94	2.42	19.8
10	Warm Temperate Forest	331.6	385.5	3.2	1.07	1.24	13.7
11	Tropical Semiarid	420.3	462.9	9.5	4.01	4.42	9.3
12	Tropical Dry Forest	452.5	504.3	14.9	6.73	7.50	10.3
13	Tropical Seasonal Forest	411.9	439.7	15.1	6.22	6.64	6.3
14	Tropical Rain Forest	333.8	345.5	8.5	2.83	2.92	3.1
TOTAL				136.7	37.84	41.50	

Aggregated Holdridge life zone is from *Leemans* [1992].

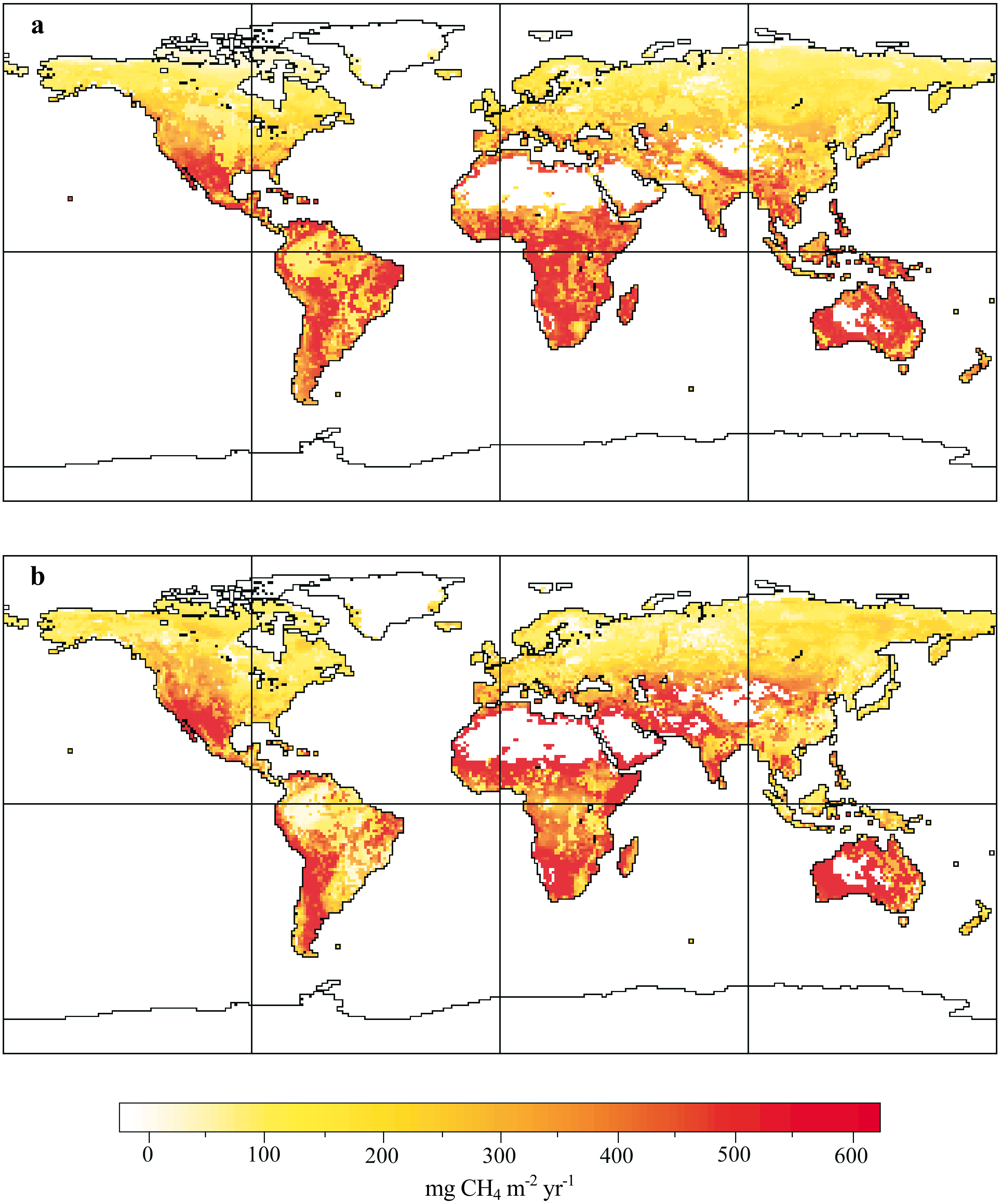


Plate 1

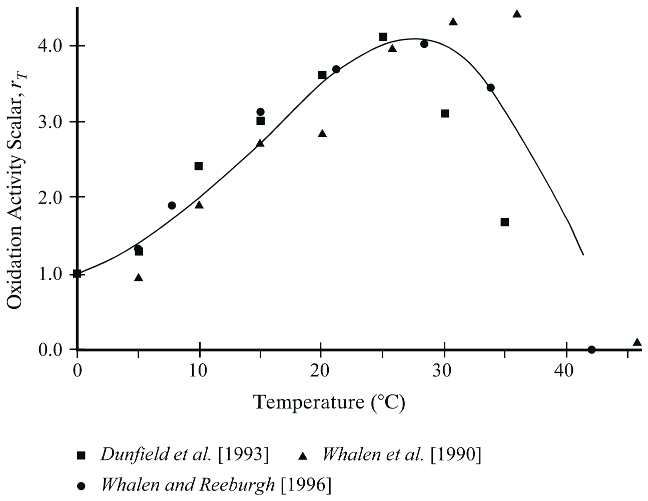

Oxidation Activity Scalar, r_T
4.0
3.0
2.0
1.0
0.0
0
10
20
30
40
Temperature (°C)
■ *Dunfield et al.* [1993]
▲ *Whalen et al.* [1990]
● *Whalen and Reeburgh* [1996]

Figure 1

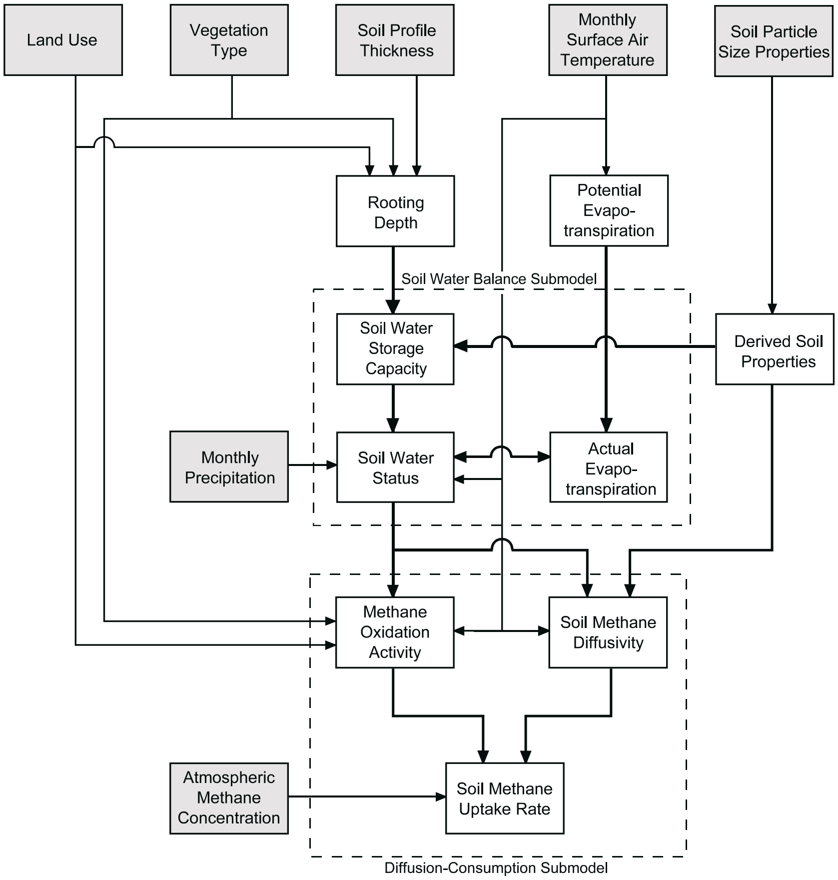

Land Use
Vegetation Type
Soil Profile Thickness
Monthly Surface Air Temperature
Soil Particle Size Properties
Rooting Depth
Potential Evapo-transpiration
Soil Water Balance Submodel
Soil Water Storage Capacity
Derived Soil Properties
Monthly Precipitation
Soil Water Status
Actual Evapo-transpiration
Methane Oxidation Activity
Soil Methane Diffusivity
Atmospheric Methane Concentration
Soil Methane Uptake Rate
Diffusion-Consumption Submodel

Figure 2

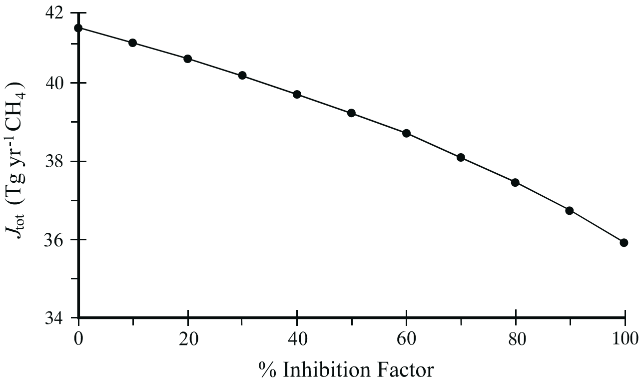
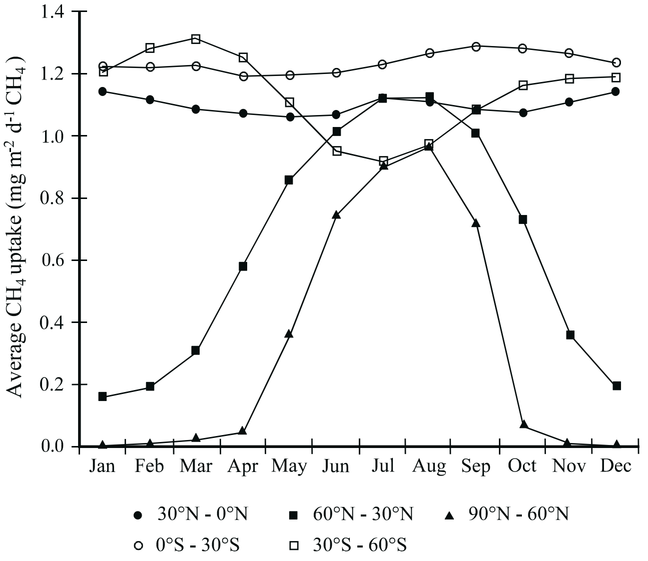

42
40
38
36
34
J_{tot} (Tg yr^{-1} CH$_4$)
0
20
40
60
80
100
% Inhibition Factor

Figure 3



Average CH4 uptake (mg m-2 d-1 CH4)
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Jan
Feb
Mar
Apr
May
Jun
Jul
Aug
Sep
Oct
Nov
Dec
● 30°N - 0°N
■ 60°N - 30°N
▲ 90°N - 60°N
○ 0°S - 30°S
□ 30°S - 60°S

Figure 4

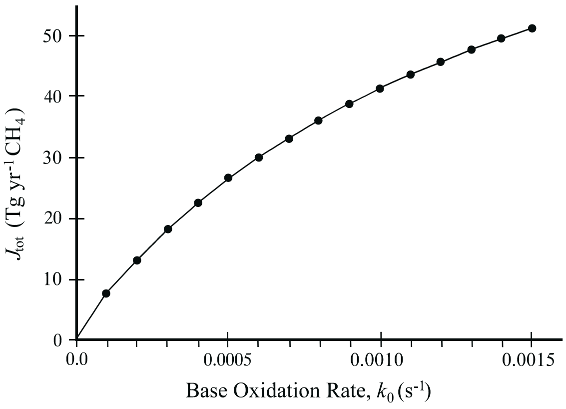
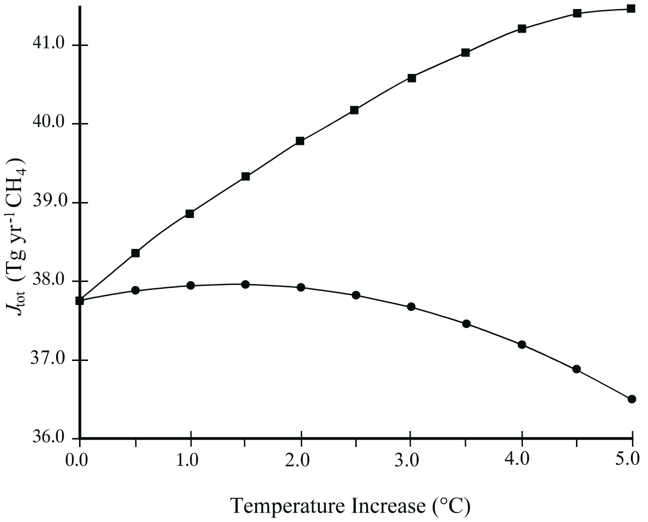

J_{tot} (Tg yr^{-1} CH$_4$)
0
10
20
30
40
50
0.0
0.0005
0.0010
0.0015
Base Oxidation Rate, k_0 (s^{-1})

Figure 5



41.0
40.0
39.0
38.0
37.0
36.0
J_{tot} (Tg yr^{-1} CH$_4$)
0.0
1.0
2.0
3.0
4.0
5.0
Temperature Increase (°C)

Figure 6